## IN THE UNITED STATES COURT OF APPEALS FOR THE FEDERAL CIRCUIT

## MAXELL, LTD., Plaintiff-Appellant

v.

## $\begin{array}{c} \text{AMPEREX TECHNOLOGY LIMITED,} \\ \textbf{\textit{Defendant-Appellee}} \end{array}$

Appeal from the United States District Court for the Western District of Texas, Case Nos. 6:21-cv-347-ADA, 6:21-cv-1007-ADA, Judge Alan D. Albright

#### OPENING BRIEF OF PLAINTIFF-APPELLANT MAXELL, LTD.

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January 30, 2023

#### Claim 1 of U.S. Patent No. 9,077,035

- 1. A nonaqueous secondary battery comprising:
- a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electrolyte,
- wherein the positive electrode comprises, as active materials, at least two lithium-containing transition metal oxides having different average particle sizes, and the lithium-containing transition metal oxide having the smallest average particle size is a lithium-containing transition metal oxide represented by the formula (1):

 $\operatorname{Li}_{\mathbf{x}} \mathbf{M}^{1}_{\mathbf{y}} \mathbf{M}^{2}_{\mathbf{z}} \mathbf{M}^{3}_{\mathbf{v}} \mathbf{O}_{2} \tag{1}$ 

wherein  $M^1$  represents at least one transition metal element selected from Co, Ni and Mn,  $M^2$  represents Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn,  $M^3$  represents at least one element selected from the group consisting of Na, K, Rb, Be, Ca, Sr, Ba, Sc, Y, La, Hf, V, Ta, Cr, Mo, W, Tc, Re, Fe, Ru, Rh, Cu, Ag, Au, B, Ca, In, Si, P and Bi, and x, y, z and v are numbers satisfying the equations respectively:  $0.97 \le x < 1.02$ ,  $0.8 \le y < 1.02$ ,  $0.002 \le z \le 0.05$ , and  $0 \le v \le 0.05$ , and has an average particle size from 2 µm to  $10 \ \mu m$ , and the lithium-containing transition metal oxide having the largest average particle size is a lithium-containing transition metal oxide represented by the formula (2):

 $Li_aM^1_bM^2_cM^3_dO_2$  (2)

wherein  $M^1$ ,  $M^2$  and  $M^3$  are the same as defined in the formula (1), and a, b, c and d are numbers satisfying the equations respectively:  $0.97 \le a < 1.02$ ,  $0.8 \le b < 1.02$ ,  $0.0002 \le c \le 0.02$ , and  $0 \le d \le 0.02$ , and has an average particle size from 5 µm to 25 µm,

wherein said electrolyte contains a fluorine-containing organic solvent, wherein the content of Co in the transition metal M<sup>1</sup> of the formulae (1) and (2) is from 30% by mole to 100% by mole,

- wherein the content of said lithium-containing transition metal oxide having the smallest average particle size in the lithium-containing transition metal oxides is from 5% by weight to 60% by weight,
- wherein the content of said lithium-containing transition metal oxide having the largest average particle size in the lithium-containing transition metal oxides is from 40% by weight to 95% by weight, and
- wherein an amount of said fluorine-containing organic solvent is 0.1% by weight to 30% by weight based on the whole weight of the electrolyte.

# UNITED STATES COURT OF APPEALS FOR THE FEDERAL CIRCUIT

#### **CERTIFICATE OF INTEREST**

Case Number	2023-1194	
<b>Short Case Caption</b>	Maxell, Ltd. v. Amperex T	echnology Limited
Filing Party/Entity	Maxell, Ltd.	
Instructions: Complete each section of the form. In answering items 2 and 3, be specific as to which represented entities the answers apply; lack of specificity may result in non-compliance. Please enter only one item per box; attach additional pages as needed and check the relevant box. Counsel must immediately file an amended Certificate of Interest if information changes. Fed. Cir. R. 47.4(b).		
I certify the following information and any attached sheets are accurate and complete to the best of my knowledge.		
Date: <u>01/30/2023</u>	_ Signature:	/s/ Hilary L. Preston
	Name:	Hilary L. Preston

1. Represented Entities. Fed. Cir. R. 47.4(a)(1).	2. Real Party in Interest. Fed. Cir. R. 47.4(a)(2).	3. Parent Corporations and Stockholders. Fed. Cir. R. 47.4(a)(3).
Provide the full names of all entities represented by undersigned counsel in this case.	Provide the full names of all real parties in interest for the entities. Do not list the real parties if they are the same as the entities.	Provide the full names of all parent corporations for the entities and all publicly held companies that own 10% or more stock in the entities.
	☑ None/Not Applicable	☑ None/Not Applicable
Maxell, Ltd.	N/A	None
	Additional pages attach	ed

<b>4. Legal Representatives.</b> List all law firms, partners, and associates that (a) appeared for the entities in the originating court or agency or (b) are expected to appear in this court for the entities. Do not include those who have already entered an appearance in this court. Fed. Cir. R. 47.4(a)(4).		
☐ None/Not Applicable	☐ Additiona	l pages attached
Paige H. Wright	Vinson & Elkins LLP	2001 Ross Ave., Suite 3900 Dallas, TX 75201
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Abigail Lubow	Unknown	712 Contra Costa Ave. Berkeley, CA 94707
<b>5. Related Cases.</b> Provide the case titles and numbers of any case known to be pending in this court or any other court or agency that will directly affect or be directly affected by this court's decision in the pending appeal. Do not include the originating case number(s) for this case. Fed. Cir. R. 47.4(a)(5). See also Fed. Cir. R. 47.5(b).		
☐ None/Not Applicable	☐ Additiona	l pages attached
Amperex Technology Limited v. Maxell, Ltd.	IPR2021-01441	PTAB
Zhuhai CosMX Battery Co., Ltd. v. Maxell, Ltd.	IPR2022-00985	PTAB
Maxell, Ltd. v. Zhuhai CosMX Battery Co., Ltd.	6:21-cv-845-ADA	W.D. Tex.
6. Organizational Victims and Bankruptcy Cases. Provide any information required under Fed. R. App. P. 26.1(b) (organizational victims in criminal cases) and 26.1(c) (bankruptcy case debtors and trustees). Fed. Cir. R. 47.4(a)(6).		
✓ None/Not Applicable	☐ Additiona	l pages attached
None		

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#### STATEMENT OF RELATED CASES

Pursuant to Federal Circuit Rule 47.5(a), counsel for Maxell, Ltd. ("Maxell") discloses that the following appeals in or from the same civil actions were previously before this Court:

- In re: Amperex Technology Ltd. (No. 2022-105): Petition for writ of mandamus denied January 14, 2022 in a per curiam order by Judges Lourie, Prost, and Taranto. In re Amperex Tech. Ltd., No. 2022-105, 2022 WL 135431 (Fed. Cir. Jan. 14, 2022).
- Amperex Technology Limited v. Maxell, Ltd. (No. 2022-1017): Appeal withdrawn following above-referenced denial of petition for writ of mandamus. Amperex Tech. Ltd. v. Maxell, Ltd., No. 2022-1017, 2022 WL 1021590 (Fed. Cir. Jan. 21, 2022).

Pursuant to Federal Circuit Rule 47.5(b), Maxell discloses that the following cases will directly affect or be directly affected by this Court's disposition of this appeal:

- Maxell, Ltd. v. Zhuhai CosMx Battery Co., Ltd., No. 6:21-cv-00845-ADA (W.D. Tex.).
- Amperex Technology Limited v. Maxell, Ltd., No. IPR2021-01441 (PTAB).
- Zhuhai CosMX Battery Co., Ltd. v. Maxell, Ltd., No. IPR2022-00985 (PTAB).

Counsel for Maxell is not aware of any other related cases within the meaning of Federal Circuit Rule 47.5.

#### JURISDICTIONAL STATEMENT

The District Court had subject-matter jurisdiction over the appealed-from civil actions pursuant to 28 U.S.C. §§ 1331 and 1338(a) because the actions—initiated by a Complaint for Declaratory Judgment of patent non-infringement, Appx1341, and a Complaint for Patent Infringement, Appx1431—arose under the patent laws of the United States, 35 U.S.C. § 1, et seq. Appx1344, Appx1434.

On November 10, 2022, the District Court entered a partial final judgment pursuant to Federal Rule of Civil Procedure 54(b), after holding that all patent claims of U.S. Patent No. 9,077,035 ("the '035 Patent") are invalid as indefinite, entering final judgment in favor of Defendant-Appellee Amperex Technology Limited on all claims involving the '035 Patent, and severing all claims involving the '035 Patent from the remaining claims pending in the case. Appx0018-0020. On November 11, 2022, Plaintiff-Appellant Maxell, Ltd. timely filed its notice of appeal.

This Court has subject-matter jurisdiction under 28 U.S.C. § 1295(a)(1) because this appeal arises from a final decision of the District Court in a civil action that arose under the patent laws of the United States, 35 U.S.C. § 1, et seq.

#### STATEMENT OF THE ISSUES

Whether the District Court erred in holding the claim term "M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole" in Claim 1 of U.S. Patent No. 9,077,035 to be indefinite as not providing a person of ordinary skill in the art with "reasonable certainty" as to whether the element cobalt is required, despite that the claim language explicitly requires at least a certain amount of the element cobalt and the specification and prosecution history confirm that cobalt is required.

#### STATEMENT OF THE CASE

#### I. Preliminary Statement

The District Court incorrectly held that the term "M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole"—hereinafter, the "Cobalt Term"—in Claim 1 of U.S. Patent No. 9,077,035 ("the '035 Patent") is indefinite and entered a partial final judgment in favor of Defendant-Appellee Amperex Technology Ltd. ("ATL") based on invalidity of the '035 Patent. Plaintiff-Appellant Maxell, Ltd. ("Maxell") respectfully requests that the Federal Circuit vacate the District Court's judgment and reverse its holding that the Cobalt Term is indefinite.

The Cobalt Term defines a portion of an active material formulation for a claimed lithium-ion battery. In analyzing this term, the District Court read the first part of the term ("M¹ represents at least one transition metal element selected from Co, Ni and Mn") as making the element cobalt ("Co") optional and the second part of the term ("wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole") as requiring cobalt. The court interpreted

that to be a contradiction that fails to give a person of ordinary skill in the art reasonable certainty as to whether cobalt is optional or required. As such, the District Court held the term to be indefinite. But the District Court's analysis was fatally flawed.

The District Court erred in its unwillingness to read the two parts of the Cobalt Term together, which resulted in its failure to recognize the claim scope defined by the intersection of the two parts of the term. When read in its entirety, a person of ordinary skill in the art would understand that the Cobalt Term clearly requires the element cobalt to be included in M<sup>1</sup>. Contrary to precedent, the District Court incorrectly read portions of the disputed claim term in isolation rather than in the context of the surrounding claim language as Section 112 requires.

Indeed, suppose a recipe for "berry pie" said "(1) the filling shall contain at least one fruit selected from blueberries, raspberries, and strawberries" and "(2) the filling must be between 30% and 100% blueberries." Any ordinary speaker of English would understand that the pie requires blueberries. To believe that blueberries might be optional, one would have to read the first part of the recipe and then stop, ignoring the rest of the recipe. Here, that is what the District Court essentially did.

Further, the District Court was wrong in construing the first part of the Cobalt Term to *require* that the elemental combinations for M<sup>1</sup> must include combinations without cobalt. Nothing in the first part of the term *requires* that M<sup>1</sup> can be just nickel ("Ni"), just manganese ("Mn"), or just a combination of nickel and manganese. It merely lists elements that can make up M<sup>1</sup>. Other limitations can further narrow the scope of M<sup>1</sup>, which is exactly what the second part of the Cobalt Term does, by adding a requirement for a minimum amount of cobalt in M<sup>1</sup>.

Thus, requiring at least 30% cobalt, while the rest of M¹ can be any combination of cobalt, nickel, or manganese, satisfies both parts of the Cobalt Term. M¹ would be "at least one transition metal element selected from Co, Ni and Mn," and "the content of Co in the transition metal M¹" would be "from 30% by mole to 100% by mole." Contrary to the District Court's analysis, there is no contradiction.

In addition, despite well-established Supreme Court and Federal Circuit precedent, the District Court did not consider the '035 Patent's specification and prosecution history in its analysis of the Cobalt Term. Had the District Court given due consideration to the intrinsic evidence, the court would have found ample confirmation that the Cobalt Term

requires the element cobalt in M<sup>1</sup>. The specification explains the importance of cobalt and provides numerous examples, all of which have at least 30% cobalt in M<sup>1</sup>. And the amendments and arguments made during prosecution, including statements explicitly relying on the required presence of cobalt to distinguish prior art, leave no room for doubt that the Cobalt Term requires cobalt in M<sup>1</sup>.

When read in view of the claim language, the specification, and the prosecution history, it is clear that the Cobalt Term requires that M¹ include cobalt. A person of ordinary skill in the art can tell if the straightforward limitations of the Cobalt Term are met or not. As such, the Cobalt Term provides greater than reasonable certainty as to the bounds of the term, and the term is not indefinite. The Court should vacate the District Court's judgment, reverse the District Court's holding that the Cobalt Term is indefinite, and hold that the Cobalt Term is to be given its plain and ordinary meaning.

II. Maxell's Attempts to Negotiate a License with ATL Lead to Lawsuits in New Jersey and Texas, and a Federal Circuit Mandamus Decision Adverse to ATL.

Maxell is a pioneer in the area of rechargeable lithium-ion batteries—the technology at issue in this appeal. See Appx1431-1433. Since

its founding in 1961, Maxell has been a leader in various industries, including energy storage and battery technologies. Appx1431. In fact, Maxell was named after its very first product, the "Maximum Capacity Dry Cell," a high-performance dry-cell battery. Id. Maxell has thus developed a significant portfolio of patents relating to battery technology, including the lithium-ion batteries that are critical to much of today's technology.

ATL is a Chinese company that produces lithium-ion batteries. On March 27, 2020, Maxell sent a letter to ATL describing Maxell's lithiumion battery patent portfolio, explaining the value of the patented technologies to ATL's current and future products, and offering to begin mutually beneficial discussions concerning ATL taking a license to Maxell's lithium-ion battery patents. Appx1433; see also Amperex Tech. Ltd. v. Maxell, Ltd. ("D.N.J. Decision"), Civ. No. 21-08461 (KM) (MF), 2021 WL 4398804, at \*1 (D.N.J. Sept. 27, 2021). The parties entered into a one-year mutual non-disclosure and litigation standstill agreement with an effective date of March 26, 2020, to facilitate negotiations. In re Amperex Tech. Ltd. ("Mandamus Denial"), No. 2022-105, 2022 WL 135431, at \*1 (Fed. Cir. Jan. 14, 2022); D.N.J. Decision, 2021 WL 4398804, at \*1.

On March 26, 2021, after a year of negotiations that failed to bear fruit and at the expiration of the non-disclosure and litigation standstill agreement, Maxell sent a letter to ATL stating that ATL's products infringed Maxell's lithium-ion battery patents and expressing Maxell's continued willingness to grant ATL a license. *D.N.J. Decision*, 2021 WL 4398804, at \*1. But the letter clearly stated that, if "Maxell and ATL are not able to enter into a licensing agreement by Friday, April 9, 2021, Maxell will be left with no choice but to pursue litigation." *Id.*; *Mandamus Denial*, 2022 WL 135431, at \*1.

After some additional communication between the parties, Maxell sent ATL an email on April 5, 2021, stating that "Maxell is open to having another meeting, but . . . request[s] that [ATL] provide the materials in advance so that [Maxell] can determine whether a written response or [a] live meeting is most appropriate." *D.N.J. Decision*, 2021 WL 4398804, at \*2. The evening of the next day, April 6, 2021, ATL's counsel responded, "I will be in touch as soon as I can get the materials." *Id.*; *Mandamus Denial*, 2022 WL 135431, at \*1.

Just over two hours after that email, ATL filed a ninety-page complaint (initiating the "New Jersey Action") in the United States District Court for the District of New Jersey (the "New Jersey Court") seeking a declaratory judgment of noninfringement of Maxell's U.S. Patent Nos. 8,691,446; 9,077,035; 9,166,251; and 9,350,019. *Mandamus Denial*, 2022 WL 135431, at \*1; *D.N.J. Decision*, 2021 WL 4398804, at \*2; *see* Appx1341. Two days later, on April 8, 2021, Maxell filed a complaint for patent infringement against ATL (initiating the "Texas Action") in the United States District Court for the Western District of Texas (the "Texas Court"), asserting the same set of patents at issue in the New Jersey Action. *Mandamus Denial*, 2022 WL 135431, at \*1; *D.N.J. Decision*, 2021 WL 4398804, at \*2; *see* Appx1431.

In the New Jersey Action, the parties filed competing motions to resolve whether the dispute would move forward in the New Jersey Court or the Texas Court. ATL filed a motion to enjoin the Texas Action under the first-to-file rule, and Maxell filed a motion to dismiss the New Jersey Action, or, in the alternative, to transfer it to the Texas Court, because ATL brought the New Jersey Action in bad faith and in anticipation of Maxell's threatened lawsuit. *Mandamus Denial*, 2022 WL 135431, at \*1; *D.N.J. Decision*, 2021 WL 4398804, at \*2. On September 27, 2021, the New Jersey Court denied ATL's injunction request and granted Maxell's

transfer request, establishing that the parties' dispute would move forward in the Texas Court. *D.N.J. Decision*, 2021 WL 4398804, at \*1, 9. The New Jersey Court concluded that "considerations of judicial and litigant economy, and the just and effective disposition of disputes require that [it] depart from the first-to-file rule." *Mandamus Denial*, 2022 WL 135431, at \*1 (quoting *D.N.J. Decision*, 2021 WL 4398804, at \*7). The fact that ATL filed the anticipatory New Jersey Action upon "receipt of specific, concrete indications" that a patent suit by Maxell was "imminent," as well as "some evidence of bad faith on ATL's behalf," along with other factors, led the New Jersey Court to exercise its discretion to decline jurisdiction and transfer the case to the Texas Court. *D.N.J. Decision*, 2021 WL 4398804, at \*5-9.

On October 4, 2021, ATL filed a notice of appeal to the Federal Circuit from the New Jersey Court's Order and Opinion denying ATL's requested injunction (docketed as Federal Circuit Case No. 2022-1017), and on October 19, 2021, ATL filed a petition for a writ of mandamus challenging the portion of the Order that transferred the New Jersey Action to Texas (docketed as Federal Circuit Case No. 2022-105). On January 14, 2022, this Court denied ATL's mandamus petition, explaining that

ATL had failed to show that the transfer ruling was a clear abuse of discretion where "the district court made a reasonable finding that ATL's suit was anticipatory" and "other considerations added to the reasons to depart from the first-to-file rule." *Mandamus Denial*, 2022 WL 135431, at \*2.

Upon the denial of its mandamus petition, ATL filed a reply brief in its pending appeal of the New Jersey Court's refusal to enjoin the Texas Action, stating that the appeal was moot in view of the mandamus outcome, which the Federal Circuit deemed a withdrawal of the appeal. *Amperex Tech. Ltd. v. Maxell, Ltd.*, No. 2022-1017, 2022 WL 1021590 (Fed. Cir. Jan. 21, 2022).

With the New Jersey Action firmly transferred to Texas, the Texas Court consolidated proceedings for the Texas Action and the New Jersey Action for all purposes, including trial. Appx1996-1997.

#### III. U.S. Patent No. 9,077,035 and the Cobalt Term

Of the four patents identified in the parties' respective complaints, this appeal relates to only one: U.S. Patent No. 9,077,035. The '035 Patent, entitled "Nonaqueous Secondary Battery and Method of Using Same," issued on July 7, 2015, from an application filed with the United

States Patent and Trademark Office on March 16, 2007, and claims priority to several Japanese patent applications dating back to March 20, 2006. Appx0119.

The '035 Patent relates to lithium-ion secondary batteries. A "secondary" battery is a rechargeable battery, while a "primary" battery is single-use and non-rechargeable. At their most basic, nonaqueous secondary batteries, including lithium-ion batteries, consist of a positive electrode, also called the cathode, and a negative electrode, also called the anode, separated from one another by a separator. Appx1747. A "nonaqueous" battery is one that immerses the positive electrode, negative electrode, and separator in a non-water-based, liquid electrolyte, which allows for the transfer of charged ions between the positive and negative electrodes through the separator. When the battery is being discharged, lithium ions move from the negative electrode (anode) through the separator to the positive electrode (cathode), and when the battery is being charged, the lithium ions move in the opposite direction. Appx1748.

The claim limitations of the '035 Patent are primarily directed to aspects of the positive electrode and the electrolyte in a nonaqueous

secondary battery. Claim 1 of the '035 Patent—the sole independent claim—reads:

- 1. A nonaqueous secondary battery comprising:
- a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electrolyte,
- wherein the positive electrode comprises, as active materials, at least two lithium-containing transition metal oxides having different average particle sizes, and the lithium-containing transition metal oxide having the smallest average particle size is a lithium-containing transition metal oxide represented by the formula (1):

$$Li_xM^1_yM^2_zM^3_vO_2 (1)$$

wherein *M¹* represents at least one transition metal element selected from *Co, Ni and Mn*, M² represents Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn, M³ represents at least one element selected from the group consisting of Na, K, Rb, Be, Ca, Sr, Ba, Sc, Y, La, Hf, V, Ta, Cr, Mo, W, Tc, Re, Fe, Ru, Rh, Cu, Ag, Au, B, Ca, In, Si, P and Bi, and x, y, z and v are numbers satisfying the equations respectively: 0.97≤x<1.02, 0.8≤y<1.02, 0.002≤z≤0.05, and 0≤v≤0.05, and has an average particle size from 2 μm to 10 μm, and the lithium-containing transition metal oxide having the largest average particle size is a lithium-containing transition metal oxide represented by the formula (2):

$$Li_aM^1_bM^2_cM^3_dO_2 (2)$$

wherein  $M^1$ ,  $M^2$  and  $M^3$  are the same as defined in the formula (1), and a, b, c and d are numbers satisfying the equations respectively:  $0.97 \le a < 1.02$ ,  $0.8 \le b < 1.02$ ,  $0.0002 \le c \le 0.02$ , and  $0 \le d \le 0.02$ , and has an average particle size from 5 µm to 25 µm,

wherein said electrolyte contains a fluorine-containing organic solvent,

- wherein the content of Co in the transition metal M<sup>1</sup> of the formulae (1) and (2) is from 30% by mole to 100% by mole,
- wherein the content of said lithium-containing transition metal oxide having the smallest average particle size in the lithium-containing transition metal oxides is from 5% by weight to 60% by weight,
- wherein the content of said lithium-containing transition metal oxide having the largest average particle size in the lithium-containing transition metal oxides is from 40% by weight to 95% by weight, and
- wherein an amount of said fluorine-containing organic solvent is 0.1% by weight to 30% by weight based on the whole weight of the electrolyte.

Appx0138 at 29:20-30:9 (emphases added). The claim term at issue in this appeal—"M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole"—appears above with emphasis. The first part of this Cobalt Term specifies the materials that M¹ may comprise in the positive electrode materials, represented by formulas 1 and 2 defined in the claim (i.e., Li<sub>x</sub>M¹<sub>y</sub>M²<sub>z</sub>M³<sub>v</sub>O₂ and Li<sub>a</sub>M¹<sub>b</sub>M²<sub>c</sub>M³<sub>d</sub>O₂, respectively): "M¹ represents at least one transition metal element selected from Co, Ni and Mn." *Id.* at 29:30-33. The second part of the term requires at least a certain amount of cobalt in M¹: "wherein the content of Co in the transition metal M¹ of

the formulae (1) and (2) is from 30% by mole to 100% by mole." *Id.* at 29:55-57.

# A. Disclosures Relating to the Cobalt Term in the '035 Patent's Specification All Include Cobalt in M<sup>1</sup>.

After describing the formulas representing the positive electrode active materials and defining the potential components of M<sup>1</sup> as "at least one transition metal element selected from Co, Ni and Mn," Appx0126 at 6:7-34, the specification describes the desirability of cobalt in M<sup>1</sup>. Specifically, the specification states:

Preferably, the proportion of Co in the lithium-containing transition metal oxide is higher, since the density of the positive electrode mixture layer can be made higher. In the formulae (1) and (2), the proportion of Co in the transition metal element M<sup>1</sup> is preferably 30% by mole or more, more preferably 65% by mole or more, particularly preferably 95% by mole or more.

#### Appx0126-0127 at 6:64-7:3.

The specification further explains that "[t]he present invention will be described in detail with reference to the following Examples." Appx0134 at 21:36-37. Every single one of the thirteen examples has cobalt in M¹ in an amount of at least 30% by mole. *See id.* at 21:48-51 (M¹ formed of cobalt alone); Appx0135 at 24:46-48 (same), 24:52-54 (same), 24:61-63 (same); Appx0136 at 25:6-11 (same), 25:18-20 (same),

25:35-37 (same), 25:51-53 (same), 25:67-26:2 (same), 26:30-32 (same), 26:41-43 (same), 26:47-49 (same); Appx0136 at 26:16 (M¹ in formula 2 approximately 33% cobalt, 33% nickel, 33% manganese); see also Appx0128 at 10:29-36 (providing formulas for positive electrode materials elsewhere in the specification, always with at least 30% cobalt in M¹).

# B. Amendments and Arguments Regarding the Cobalt Term in the '035 Patent's Prosecution History Emphasize the Requirement for Cobalt in M<sup>1</sup>.

As originally filed, the application that led to the '035 Patent required that for all claims, "M¹ represents at least one transition metal element selected from Co, Ni and Mn."¹ Appx0214. Early in prosecution, the examiner issued an election/restriction office action, requiring Maxell to "[p]lease define M1 as x) Co or y) Ni or z) Mn [please pick one from (x)-(z)]." Appx0511 (second bracketed text and emphasis in original). In response, Maxell elected that "in the formula Li<sub>a</sub>M¹<sub>b</sub>M²<sub>c</sub>M³<sub>d</sub>O₂ M¹ is x) CO." Appx0517 (emphases in original). This election established the importance and necessity of cobalt in the material M¹.

<sup>&</sup>lt;sup>1</sup> The language appeared in original Claim 3 filed with the application, Appx0213-0214, which was later merged into Claim 1, Appx1209; Appx1214.

Later claim amendments during prosecution, and Maxell's attendant explanations, established that cobalt was a necessary component of M<sup>1</sup>. Specifically, on May 19, 2014, Maxell amended Claim 1, in relevant part, to read:

wherein M<sup>1</sup> represents at least one transition metal element selected from Co, Ni and Mn, . . . .

Appx1251; see Appx1259. This amendment thus specified that  $M^1$  was made up **solely** of cobalt. Distinguishing a prior art reference relied upon by the examiner, Oda et al. (JP 2005-302507) ("Oda"), Maxell relied on this composition of  $M^1$ , explaining that the Oda reference "does <u>not</u> describe or teach that the transition metal consists of Co alone, since [Oda] predominantly uses Ni as a transition metal." Appx1260 (emphasis in original).

Shortly thereafter, on June 19, 2014, in reply to an examiner's advisory action indicating that the prior amendments still failed to place the application in condition for allowance, Appx1267, Maxell again amended Claim 1, in relevant part, as follows:

wherein  $M^1$  represents <u>at least one transition metal elements</u> selected from Co, Ni and Mn, . . .

. . .

## wherein the content of Co in the transition metal $M^1$ is 30% by mole or more.

Appx1272-1273. Although the new amendment allowed M¹ to also contain nickel or manganese, the addition of the content requirement for cobalt—at the same time—ensured that cobalt remained required. Then, Maxell again went on to distinguish the prior art based on the required presence of cobalt in the newly amended claim. Emphasizing precisely the two amended limitations quoted above, see Appx1279-1280, Maxell once more distinguished the Oda reference, by arguing that "[Oda] does not describe or teach such a content of Co, since [Oda] predominantly uses Ni as a transition metal," Appx1280 (bold emphasis added).

The examiner clearly agreed, because in the next office action, the examiner dropped all prior art-based rejections. See Appx1293-1299. The examiner understood that cobalt was a necessary component of  $M^1$  and was able to apply that understanding in order to withdraw her prior rejection based on Oda.

After dropping all prior art-based rejections, the examiner introduced a number of Section 112 objections, including two addressed to the presence of cobalt in M<sup>1</sup>. Appx1295-1298. Specifically, the examiner raised the following rejections:

10. Claim[] 1 . . . [is] rejected under 35 U.S.C. 112(a) or 35 U.S.C. 112 (pre-AIA), first paragraph, as based on a disclosure which is not enabling. The upper range of the content of the Co in the transition metal M1 is critical or essential to the practice of the invention . . . .

. . .

13. Claim[] 1 . . . [is] rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite . . . .

In claim 1 it is unclear which M1, Formula (1) or Formula (2) is being discussed in the phrase "wherein the content of Co in the transition metal M1 is 30% by mole".

Appx1297-1298. The examiner did *not* issue an indefiniteness rejection based on any lack of clarity as to whether the claims require cobalt in M<sup>1</sup>. To the contrary, the enablement rejection specifies that the "upper range of the *content of the Co* in the transition metal M1 is *critical or essential* to the practice of the invention," indicating that the examiner understood that some content of cobalt is necessary in M<sup>1</sup>. Appx1297 (emphases added). Similarly, the indefiniteness rejection was based on a lack of specificity regarding which formula required the presence of cobalt, thus presupposing the necessary presence of cobalt. Appx1298.

In response, Maxell amended Claim 1, giving the Cobalt Term its current form:

wherein  $M^1$  represents at least one transition metal element selected from Co, Ni and Mn, . . .

. . .

wherein the content of Co in the transition metal  $M^1$  of the formulae (1) and (2) is from 30% by mole or more to 100% by mole, . . . .

Appx1303-1304. The examiner accepted that these amendments resolved the Section 112 rejections and issued a notice of allowance. Appx1320, Appx1325.

# IV. The District Court Holds the Cobalt Term Is Indefinite and Issues a Partial Final Judgment.

During claim construction, ATL argued that the Cobalt Term is indefinite, and the District Court ultimately agreed. Appx0025; Appx0068-0072. ATL contended that the term is indefinite because "it is unclear whether Co has to be selected or is merely optional." Appx0068. Specifically, ATL argued "that the first part of the claim describes that Co is optional while the second half of the claim recites that Co is required," and that "these conflicting requirements" render the term indefinite. *Id.* Maxell rebutted ATL's arguments, explaining how the Cobalt Term does not constitute a contradiction and how the '035 Patent's intrinsic record supports the clear requirement for cobalt to be included in M¹. Appx0069.

Following the completion of briefing, on the eve of the February 14, 2022 Markman hearing, the District Court sent the parties its preliminary constructions, including a preliminary construction that the Cobalt Term is indefinite. During the *Markman* hearing, Maxell argued as to why ATL and the preliminary construction were wrong, again explaining the lack of a contradiction and walking the court through the '035 Patent's specification and prosecution history to show that a person of ordinary skill would clearly understand that M1 must include cobalt. Appx2204-2211. Without hearing any argument from ATL, and without any explanation as to its reasoning, the District Court held that it would "maintain its construction of indefinite" for the Cobalt Term. Appx2211. That same day, the District Court issued its Claim Construction Order, comprising a table showing that the Cobalt Term was indefinite, but without providing any further reasoning. Appx0025.

On March 9, 2022, Maxell filed a motion seeking reconsideration of the District Court's indefiniteness holding, or alternatively, for entry of partial final judgment under Federal Rule of Civil Procedure 54(b). Appx0095. The motion sought to convince the District Court to change its holding that the Cobalt Term is indefinite by arguing that the clear language of the claim, the specification, and the prosecution history all leave no doubt that the element cobalt must be included in M<sup>1</sup>. In the alternative, with the indefiniteness holding effectively rendering all claims of the '035 Patent invalid, Maxell sought entry of partial final judgment as to the '035 Patent so that it could be severed from the remaining patents in order to allow Maxell to pursue an appeal without delay.

The District Court held oral argument on Maxell's motion on July 27, 2022. Appx2249, Appx2251-2281, Appx2287. After hearing from both sides on Maxell's motion for reconsideration, the court stated it was "maintain[ing] its construction of indefinite" for the Cobalt Term. Appx2271. Following additional argument on the alternative motion for entry of partial final judgment, the court stated that it would "prepare as quickly as possible an order explaining the reasons for finding the claim term indefinite," and once that was entered, the court would "enter a partial summary judgment with respect to that patent on that basis" and "sever that issue" with "a judgment that will be appealable." Appx2287.

On November 10, 2022, the District Court issued a Claim Construction Order and Memorandum in Support Thereof, providing, *inter alia*, the court's analysis as to why it held the Cobalt Term indefinite. Appx0068-0072; see Maxell, Ltd. v. Amperex Tech. Ltd., No. W-21-CV-00347-ADA, 2022 WL 16858824, at \*19-21 (W.D. Tex. Nov. 10, 2022). With regard to the Cobalt Term, after summarizing the parties' positions, the District Court provided four reasons why it found the term indefinite:

- First, the court said "the plain language of the claim recites a contradiction" because the first part of the Cobalt Term makes cobalt optional while the second part makes it required, and "[f]or an element to simultaneously be optional and required is a contradiction on its face." Appx0070.
- Second, the court disagreed with Maxell's argument that, reading both parts of the term together, M¹ could be (1) cobalt, or (2) cobalt plus nickel and/or manganese, because the court interpreted the first part of the Cobalt Term to mean that M¹ can be any combination of cobalt, nickel, or manganese, with some combinations containing no cobalt at all. Appx0071.
- Third, the court disagreed with Maxell's argument that, following standard patent drafting practice, the second part of the Cobalt Term simply narrowed the broader first part, because "the

second part of the claim does not narrow the first part of the claim as it does not apply to some [combinations of  $M^1$  elements]." Id.

• Fourth, the court found that if Maxell had wanted to make cobalt required, it knew how, based on claim language in an unrelated patent also being asserted against ATL that required "Mg, or Mg and at least [one other element selected from a list]." Appx0071-0072.

The District Court thus held that the Cobalt Term is indefinite. Appx0072. The court did not make any underlying findings of fact, but instead based its holding on the claim language alone. Further, although the court acknowledged that Maxell made arguments based on the intrinsic record in its summary of Maxell's position, the court did not refer to either the specification or the prosecution history at all in its analysis. See Appx0070-0072.

Also on November 10, 2022, the District Court issued a Partial Final Judgment Pursuant to Rule 54(b). Appx0018-0020. Based on the court's holding that the Cobalt Term is indefinite, the court granted summary judgment that all claims of the '035 Patent are invalid and issued

a final judgment in ATL's favor with respect to all claims and counterclaims involving the '035 Patent, severing the '035 Patent from the case. Appx0018-0019. The District Court found that there was no just reason for delay and certified the judgment concerning the '035 Patent as final pursuant to Rule 54(b). Appx0020.

Maxell timely appealed and requests that the Federal Circuit vacate the District Court's judgment, reverse the District Court's holding that the Cobalt Term is indefinite, and hold that the Cobalt Term should be given its plain and ordinary meaning.

### **SUMMARY OF THE ARGUMENT**

The District Court incorrectly held that the term "M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole"—the Cobalt Term—in Claim 1 of U.S. Patent No. 9,077,035 is indefinite, and this Court should reverse that holding.

The District Court read the first part of the term as making the element cobalt optional and the second part of the term as making cobalt required, and interpreted that to be a contradiction that fails to give a person of ordinary skill in the art reasonable certainty as to whether cobalt is optional or required. However, the first part of the term does not require that the elemental combinations for M<sup>1</sup> must include combinations without cobalt; it merely lists elements that can make up M<sup>1</sup>. Other limitations can further narrow the scope of M<sup>1</sup>, which is exactly what the second part of the Cobalt Term does by adding a requirement for a minimum amount of cobalt in M<sup>1</sup>. Thus, when read in its entirety, a person of ordinary skill in the art would understand that the Cobalt Term clearly requires the element cobalt to be included in M<sup>1</sup>.

Indeed, the Cobalt Term does not contain a contradiction as the District Court held, because it is possible to satisfy both parts of the term by having at least 30% cobalt in M¹, while the rest of M¹ can be any combination of cobalt, nickel, or manganese. Under those circumstances, M¹ would be "at least one transition metal element selected from Co, Ni and Mn," and "the content of Co in the transition metal M¹" would be "from 30% by mole to 100% by mole." A person of ordinary skill in the art can tell if the straightforward limitations of the Cobalt Term are met or not. As such, the Cobalt Term provides greater than reasonable certainty as to the bounds of the term, and the term is not indefinite.

The '035 Patent's specification and prosecution history—which Supreme Court and Federal Circuit precedent hold must be considered when analyzing indefiniteness—confirm that the Cobalt Term requires the element cobalt to be present in M¹. However, the District Court failed to address any of this intrinsic evidence in its analysis. The specification explains the importance of cobalt and provides numerous examples of positive electrode materials according to the claimed invention, all of which have at least 30% cobalt in M¹, consistent with the claim limitations. And the amendments and arguments made during prosecution,

including statements explicitly relying on the required presence of cobalt to distinguish prior art, leave no room for doubt that the Cobalt Term requires cobalt in  $M^1$ .

The District Court misconstrued the Cobalt Term to have a contradiction where there is none and incorrectly held the term indefinite as a result. But the correct reading of the term, in view of the claim language, the specification, and the prosecution history, confirms that M¹ must include cobalt. Because the District Court granted judgment in ATL's favor based on its erroneous holding that the Cobalt Term is indefinite, the Federal Circuit should vacate the District Court's judgment and reverse the indefiniteness holding. As the language of the Cobalt Term is clear, the term should be given its plain and ordinary meaning.

#### **ARGUMENT**

#### I. Standard of Review

"Claim indefiniteness is a legal conclusion, in implementation of 35 U.S.C. § 112." Nature Simulation Sys. Inc. v. Autodesk, Inc., 50 F.4th 1358, 1360 (Fed. Cir. 2022). "Indefiniteness, therefore, like claim construction, is a question of law that [the Court] review[s] de novo." Atmel Corp. v. Info. Storage Devices, Inc., 198 F.3d 1374, 1378 (Fed. Cir. 1999). "As in claim construction, [the Court] review[s] a district court's underlying factual determinations for clear error." Cox Commc'ns, Inc. v. Sprint Commc'n Co., 838 F.3d 1224, 1228 (Fed. Cir. 2016). Here, however, the District Court did not make any factual findings and based its holding on the language of the claim only. "If indefiniteness can be determined based solely on intrinsic evidence, [the Court's] review is de novo." Id. "Indefiniteness must be proven by clear and convincing evidence." Sonix Tech. Co. v. Publ'ns Int'l, Ltd., 844 F.3d 1370, 1377 (Fed. Cir. 2017).

II. The Law Requires Patent Claims to Provide Reasonable Certainty as to the Scope of an Invention to Define the Boundaries of the Right to Exclude for the Public.

The issue presented in this appeal is whether the Cobalt Term is indefinite. "Patent claims must provide reasonable certainty in defining

what is patented, in conformity with the requirements of 35 U.S.C. § 112." *Nature Simulation Sys.*, 50 F.4th at 1362. The second paragraph of Section 112 requires:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

35 U.S.C. § 112 ¶ 2 (pre-AIA).² In Nautilus, Inc. v. Biosig Instruments, Inc., the Supreme Court held that in order to satisfy this requirement, patent claims, "viewed in light of the specification and prosecution history, [must] inform those skilled in the art about the scope of the invention with reasonable certainty." 572 U.S. 898, 910 (2014). Although the Court recognized that "absolute precision is unattainable," it explained that "a patent must be precise enough to afford clear notice of what is claimed, thereby 'appris[ing] the public of what is still open to them." Id. at 909, 910 (quoting Markman v. Westview Instruments, Inc., 517 U.S. 370, 373 (1996)) (quotation marks omitted). Thus, the purpose of the definiteness requirement is to define the bounds of an invention, so that it

<sup>&</sup>lt;sup>2</sup> The pre-AIA version of Section 112 applies to the '035 Patent because the patent has an effective date prior to September 16, 2012. In any event, the current AIA version of Section 112(b) uses substantially the same language as the second paragraph of pre-AIA Section 112.

is possible to determine what falls within the zone of exclusion and what remains to the public:

[The claims'] principal function, therefore, is to provide notice of the boundaries of the right to exclude and to define limits; it is not to describe the invention, although their original language contributes to the description and in certain cases satisfies it. Claims define and circumscribe, the written description discloses and teaches.

Nature Simulation Sys., 50 F.4th at 1363 (quoting Ariad Pharms., Inc. v. Eli Lilly & Co., 598 F.3d 1336, 1347 (Fed. Cir. 2010)).

Further, as the Supreme Court stated in *Nautilus*, the claims must be "viewed in light of the specification and prosecution history." 572 U.S. at 910. Here, the District Court held the Cobalt Term indefinite based on the claim language alone, but "[c]laim language, standing alone,' is not the correct standard of law and is contrary to uniform precedent." *Nature Simulation Sys.*, 50 F.4th at 1364. "Patent claims are viewed and understood in light of the specification, the prosecution history, and other relevant evidence, as 'would have allowed a skilled artisan to know the scope of the claimed invention with reasonable certainty." *Id.* (quoting *Sonix*, 844 F.3d at 1376).

As such, if a person of ordinary skill in the art ("POSITA") can determine with reasonable certainty whether something falls inside or

outside the scope of the claims, when viewed in light of the specification and prosecution history, then a claim is not indefinite. Here, the Cobalt Term meets this standard because it is clear that cobalt is required, so the term is therefore not indefinite.

# III. The Cobalt Term Clearly Defines the Bounds of the Invention and Informs a Person of Skill in the Art that the Element Cobalt Is Required.

When read in its entirety, a person of ordinary skill in the art would understand that the Cobalt Term clearly requires the element cobalt to be included in M<sup>1</sup>. Indeed, given the straightforward language of the Cobalt Term, a POSITA would have no trouble discerning whether something falls inside or outside the bounds of the '035 Patent's claims. As such, the Cobalt Term is not indefinite.

"We begin our [indefiniteness] analysis with the claim language." *Tinnus Enters.*, *LLC v. Telebrands Corp.*, 733 F. App'x 1011, 1018 (Fed. Cir. 2018). That analysis must encompass the claim as a whole, because it is error to "read the disputed claim term in isolation rather than in the context of the surrounding claim language as § 112(b) requires." *Id.* at 1020. Here, the Cobalt Term contains two parts:

[1] M¹ represents at least one transition metal element selected from Co, Ni and Mn,

and

[2] wherein the content of Co in the transition metal M<sup>1</sup> of the formulae (1) and (2) is from 30% by mole to 100% by mole[.]

Appx0138 at 29:32-33, 29:55-57; see Appx0068. The District Court's analysis noted that "[t]he first part of the claim recites a Markush group where Co is not necessarily required to be in the claimed compound while the second part of the claim recites that Co is necessarily required." Appx0070. If looking at each part of the Cobalt Term in isolation was appropriate, the District Court's description would not be wrong. However, the District Court erred because it failed to read the two parts of the Cobalt Term together and thus recognize the claim scope defined by the intersection of the two parts of the term. See Tinnus, 733 F. App'x at 1020 (relying on the "context of the surrounding claim language" to "further define]" limitations).

### A. The Second Part of the Cobalt Term Narrows the Composition of M<sup>1</sup> from the Broader First Part.

The first part of the Cobalt Term establishes the potential metals that can form M¹: cobalt, nickel, and manganese. If the claim ended there, M¹ could be any one of those three metals alone, or any combination of those three metals. But the claim does not end there; the second part of the term states that cobalt must be present in M¹ in an amount of

30% to 100% by mole. The language of the second part is mandatory, not optional. It clearly and unmistakably narrows the scope of the claim by requiring the presence of cobalt, such that M¹ can be cobalt alone, or a combination of cobalt with nickel and/or manganese, provided that the proportion of cobalt is at least 30%. There is no "zone of uncertainty," against which the Supreme Court has warned, because there is no question that cobalt is required. *See Nautilus*, 572 U.S. at 911 (quotation marks omitted).

The District Court erred in concluding that "the plain language of the claim recites a contradiction" and that the Cobalt Term constitutes a "contradiction on its face" because cobalt is "simultaneously . . . optional and required." Appx0070. This is **not** a case where a claim is indefinite because two limitations present an irreconcilable contradiction. See, e.g., Multilayer Stretch Cling Film Holdings, Inc. v. Berry Plastics Corp., 831 F.3d 1350, 1362 (Fed. Cir. 2016) ("Independent claim 1 excludes LDPE from the inner layers, while dependent claim 10 includes it. As such, claim 10 is inconsistent with claim 1 and, indeed, contradicts claim 1."); Trs. of Columbia Univ. in City of N.Y. v. Symantec Corp., 811 F.3d 1359, 1367 (Fed. Cir. 2016) ("The claims are nonsensical in the way a claim to

extracting orange juice from apples would be, and are thus indefinite."). Rather, it is a simple matter to satisfy both parts of the Cobalt Term. Indeed, the *only* way to satisfy both parts of the term is to require the presence of cobalt in M¹. Such a reading of the term is both consistent with the intrinsic record and also preserves the validity of the '035 Patent's claims. *See Modine Mfg. Co. v. U.S. Int'l Trade Comm'n*, 75 F.3d 1545, 1557 (Fed. Cir. 1996) (rejecting indefiniteness argument and stating that "[w]hen claims are amenable to more than one construction, they should when reasonably possible be interpreted so as to preserve their validity"), abrogated on other grounds by Festo Corp. v. Shoketsu Kinzoku Kogyo Kabushiki Co., 234 F.3d 558 (Fed. Cir. 2000) (en banc).³

As Maxell explained to the District Court, when both parts of the Cobalt Term are read *together*, the resulting elemental combinations for M<sup>1</sup> are essentially "[1] Co, or [2] Co and at least one transition metal

<sup>&</sup>lt;sup>3</sup> See also Brown v. Guild, 90 U.S. (23 Wall.) 181, 226 (1874) ("[W]e ought to give a favorable construction, so as to sustain the patent if it can fairly be done."); Klien v. Russell, 86 U.S. (19 Wall.) 433, 466 (1873) ("The court should proceed in a liberal spirit, so as to sustain the patent and the construction claimed by the patentee himself, if this can be done consistently with the language which he has employed."); Turrill v. Mich. S. & N. Ind. R.R. Co., 68 U.S. (1 Wall.) 491, 510 (1863) ("Patents for inventions . . . are to receive a liberal construction, and . . . are, if practicable, to be so interpreted as to uphold and not to destroy the rights of the inventor.").

element selected from the group consisting of Ni and Mn." See Appx0071. The court incorrectly rejected this argument by looking only at the first part of the Cobalt Term and concluding that the claim permits M<sup>1</sup> to be any of "up to seven options (Co, Ni, Mn, Ni & Mn, Co & Mn, Co & Ni, Co & Ni & Mn)." Id. However, although the first part of the term lists three possible elements—cobalt, nickel, or manganese—nothing in the term requires that every possible combination of those elements—including combinations lacking cobalt—must be available in the context of the rest of the claim. Instead, the limitation "M1 represents at least one transition metal element selected from Co, Ni and Mn" is satisfied simply if M<sup>1</sup> is one or a combination of those elements, even if in every case M<sup>1</sup> includes cobalt. In other words, nothing in the first part of the term requires that M<sup>1</sup> can be just nickel, just manganese, or just a combination of nickel and manganese, regardless of the remaining limitations. The District Court's reasoning incorrectly imports such a requirement into the first part of the term. See Appx0070-0071.

The District Court also erred when it agreed with ATL that had Maxell intended to require cobalt in M<sup>1</sup>, it "knew how to do so" based on different claim language from U.S. Patent No. 9,350,019 ("the '019

Patent"). See Appx0071-0072. The '019 Patent, also asserted by Maxell against ATL, may share many characteristics with the '035 Patent, but the '019 Patent is not in the same family as the '035 Patent and did not share the same patent examiner. Compare Appx0119 with Appx1375. It is therefore extrinsic to the issue of interpreting the terms of the '035 Patent. In any event, the District Court cited no authority, and Maxell is not aware of any, that requires a patent to claim the necessary presence of an element using any particular language. The relevant question here is whether the "claims, read in light of the specification delineating" the patent, and the prosecution history, . . . inform, with reasonable certainty, those skilled in the art about the scope of the invention." Nautilus, 572 U.S. at 901. As shown herein, the Cobalt Term clearly exceeds this standard based on the intrinsic record.

## B. A POSITA Can Identify the Bounds of the Cobalt Term with Reasonable Certainty.

Because the first part of the Cobalt Term does not require that the elemental combinations for M<sup>1</sup> must include combinations without cobalt, it is a straightforward matter for a POSITA to determine whether a formulation of M<sup>1</sup> falls within the scope of the Cobalt Term. For example, cobalt alone, or 50% cobalt and 50% nickel, satisfies both limitations. A

combination of 25% cobalt and 75% manganese would satisfy the first part but not the second part of the Cobalt Term—because there is not at least 30% cobalt—and would fall outside of the '035 Patent's claims. Likewise, a combination of just nickel and manganese would satisfy the first part of the term but would not satisfy the second—again because there is not at least 30% cobalt—so that would also fall outside the scope of the '035 Patent.

When viewed in its entirety and giving effect to both parts of the term, the boundaries of the Cobalt Term are clear to a POSITA. The parties here appear to agree that someone with a bachelor's degree in chemistry or chemical engineering and multiple years of experience with lithium-ion batteries would qualify as a POSITA. See Appx1744-1745; Appx1766. It is "difficult to believe" that such a POSITA "who had read the specification and relevant prosecution history would be unable to determine with reasonable certainty" whether cobalt is required and in what quantities. See Tinnus, 733 F. App'x at 1015 (internal quotations omitted).

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<sup>&</sup>lt;sup>4</sup> The level of ordinary skill in the art was not addressed in detail during claim construction, so the District Court did not issue a finding regarding the definition of a POSITA.

Indeed, both parties' experts here understood how to determine whether a formulation for M<sup>1</sup> satisfies both parts of the Cobalt Term. Maxell's expert, Dr. Brett Lucht, explained "that Co is a required element that must be present in M<sup>1</sup>, and that it can comprise as much as 100% or as little as 30% of M<sup>1</sup>. If M<sup>1</sup> is comprised of less than 100% Co, the remaining content of M<sup>1</sup> must be comprised of Ni or Mn." Appx1778. Likewise, ATL's expert, Dr. Thomas Fuller, recognized that "the patent indicates that cobalt must be between 30% and 100% of M<sup>1</sup>." Appx1753. He was further able to identify how formulations of M<sup>1</sup> could satisfy one part of the term but not the other, and fall outside the claim scope, noting that "multiple cobalt-containing alloys that are manifestly within the phrase 'selected from Co, Ni and Mn'—for example, 20% Co, 40% Ni, and 40% Mn—are excluded by the later limitation that 'the content of Co' in M<sup>1</sup> 'is from 30% by mole to 100% by mole." Id. The District Court did not address this confirmatory expert testimony in its order. See Appx0068-0072.

"The purpose of claims is not to explain the technology or how it works, but to state the legal boundaries of the patent grant." S3 Inc. v. NVIDIA Corp., 259 F.3d 1364, 1369 (Fed. Cir. 2001); see also Nature

Simulation Sys., 50 F.4th at 1362 ("[The claims'] principal function . . . is to provide notice of the boundaries of the right to exclude and to define limits . . . . Claims define and circumscribe . . . ." (quoting Ariad Pharms., 598 F.3d at 1347)). Here, because a POSITA can easily determine whether a formulation of M¹ satisfies the two parts of the Cobalt Term, as both parties' experts readily did, the term defines the boundaries of the claimed invention with reasonable certainty and is not indefinite. Nautilus, 572 U.S. at 909-10.

### C. Narrowing Limitations Such as the Second Part of the Cobalt Term Are a Standard Claim Drafting Practice.

As explained above, the second part of the Cobalt Term narrows the scope for the composition of M¹ from the broader first part of the term. Such narrowing limitations are a standard practice in claim drafting. See, e.g., Multilayer Stretch, 831 F.3d at 1354 (construing claim with "five inner layers" defined by Markush group narrowed by later limitation requiring one layer to be a specific identified material with specific characteristics). The Markush group of cobalt, nickel, and manganese provides a broad scope that begins to define M¹, and the additional limitation that M¹ must contain at least 30% cobalt provides further narrowing definition to M¹. The District Court disagreed that the second part narrows

the first part, reasoning that "the second part of the claim does not narrow the first part of the claim as it does not apply to some Markush group combinations." Appx0071. There, the District Court appeared to say that since the second part of the Cobalt Term specifies a certain amount of cobalt, it cannot narrow elemental combinations that do not contain cobalt. But that incorrectly assumes that  $M^1$  must be able to have elemental combinations that do not contain cobalt. What the District Court failed to understand is that requiring a certain amount of cobalt necessarily narrows the claim by removing elemental combinations that do not contain cobalt from the scope of  $M^1$ .

Indeed, a more extreme version of the narrowing limitation found in the second part of the Cobalt Term can be seen in Claim 10 of the '035 Patent, which depends from Claim 1 and requires simply that "M¹ represents Co." Appx0138 at 30:52-53; see also Appx1276, Appx1280 (adding dependent claim during prosecution and arguing its requirement for cobalt, at the same time that the separate 30% requirement for cobalt was added to Claim 1). Claim 10 narrows the scope of M¹ to only cobalt, but under the District Court's reasoning, it would also be indefinite because it "contradicts" rather than narrows the first part of the Cobalt Term

supposedly allowing M¹ to be any combination of cobalt, nickel, or manganese. Surely that would be the wrong result, as a POSITA would know from the dependent limitation that M¹ must be cobalt alone. The Cobalt Term is no different, as that term and Claim 10 both do the same thing: narrow the scope of M¹ by requiring a certain amount of cobalt—at least 30% cobalt and 100% cobalt, respectively.

The fact that the limitation in Claim 10 appears in a dependent claim is a distinction without a difference. "A claim in dependent form shall be construed to incorporate by reference all the limitations of the claim to which it refers." 35 U.S.C. § 112 ¶ 4 (pre-AIA); see also Honeywell Int'l Inc. v. Hamilton Sundstrand Corp., 370 F.3d 1131, 1142 (Fed. Cir. 2004) (en banc) (noting that when rewriting a dependent claim into independent form, "the scope of the rewritten claim has remained unchanged"). Thus, Claim 10 could be rewritten to put the limitations "M¹ represents at least one transition metal element selected from Co, Ni and Mn" and "M¹ represents Co" together in the same claim. This rewritten claim language would not be that different from the Cobalt Term. If the rewritten claim language has identical scope to Claim 10 and is no less

definite than Claim 10, then it must hold true that the very similar Cobalt Term also cannot be indefinite.

Because the plain language of the Cobalt Term defines the bounds of the invention to a POSITA with beyond reasonable certainty, the Cobalt Term is not indefinite by clear and convincing evidence, and the term should be given its plain and ordinary meaning.

### IV. The Specification of the '035 Patent Clearly Supports the Necessary Presence of Cobalt in M<sup>1</sup>.

Although, as demonstrated above, the plain language of the Cobalt Term delineates the scope of the invention with even greater than reasonable certainty, if a POSITA were still unsure whether the term requires M¹ to contain cobalt, then the review turns to "the specification delineating the patent," which additionally supports the need for cobalt. Nautilus, 572 U.S. at 901. Despite Supreme Court and Federal Circuit dictates, the District Court did not address the specification at all in its indefiniteness analysis. See Appx0070-0072. "As always, whether a claim is indefinite must be judged in light of the specification . . .' of the patent in which it appears." Sonix, 844 F.3d at 1381 (quoting Interval Licensing LLC v. AOL, Inc., 766 F.3d 1364, 1369 (Fed. Cir. 2014)); see also Nautilus, 572 U.S. at 901 (holding claims must be "read in light of

the specification delineating the patent"). Here, the specification of the '035 Patent clearly supports the necessary presence of cobalt in M¹.

After describing the formulas representing the positive electrode active materials and defining the potential components of M<sup>1</sup> as "at least one transition metal element selected from Co, Ni and Mn," Appx0126 at 6:7-34, the specification further describes the desirability of cobalt in M<sup>1</sup>. Specifically, the specification states:

Preferably, the proportion of Co in the lithium-containing transition metal oxide is higher, since the density of the positive electrode mixture layer can be made higher. In the formulae (1) and (2), the proportion of Co in the transition metal element M<sup>1</sup> is preferably 30% by mole or more, more preferably 65% by mole or more, particularly preferably 95% by mole or more.

Appx0126-0127 at 6:64-7:3. That is, the specification clearly describes the importance of cobalt and identifies exactly the content requirement recited in the Cobalt Term.

Beyond this guidance, the specification further notes that "[t]he present invention will be described in detail with reference to the following [thirteen] Examples." Appx0134 at 21:36-37. Of note, every single one of the thirteen examples is consistent with the teachings of the specification and the requirements of the Cobalt Term—cobalt is present in

every single example, at least 30% by mole. *See id.* at 21:48-51 (M¹ formed of cobalt alone); Appx0135 at 24:46-48 (same), 24:52-54 (same), 24:61-63 (same); Appx0136 at 25:6-11 (same), 25:18-20 (same), 25:35-37 (same), 25:51-53 (same), 25:67-26:2 (same), 26:30-32 (same), 26:41-43 (same), 26:47-49 (same); Appx0136 at 26:16 (M¹ in formula 2 approximately 33% cobalt, 33% nickel, 33% manganese); *see also* Appx0128 at 10:29-36 (providing formulas for positive electrode materials elsewhere in the specification, always with at least 30% cobalt in M¹).

In light of these teachings of the specification, which must be considered pursuant to Supreme Court and Federal Circuit precedent, a POSITA reading the Cobalt Term would have beyond reasonable certainty regarding the necessary presence of cobalt in M<sup>1</sup>.

## V. The Prosecution History of the '035 Patent Clearly and Unmistakably Shows that Cobalt Is Required.

Should the plain language of the claim itself and the clear teachings of the specification still leave a POSITA uncertain as to the scope of the Cobalt Term—which they should not—the prosecution history leaves no doubt that the element cobalt is required in M<sup>1</sup>. Just as with the specification, the District Court did not address the prosecution history at all in its analysis concluding that the Cobalt Term is indefinite, Appx0070-

0072, despite acknowledging that Maxell presented arguments regarding the prosecution history, Appx0069. This was clear error, as "[c]laim language, standing alone' is not the correct standard of law and is contrary to uniform precedent." *Nature Simulation Sys.*, 50 F.4th at 1364. As the Supreme Court and Federal Circuit have uniformly held, "in assessing definiteness, claims are to be read in light of the patent's . . . prosecution history." *Nautilus*, 572 U.S. at 908; *see also Ancora Techs.*, *Inc. v. Apple, Inc.*, 744 F.3d 732, 738 (Fed. Cir. 2014) ("[A]n ordinarily skilled artisan must consult the prosecution history to confirm the proper understanding of a claim term's meaning, especially if other aspects of the inquiry raise questions.").

## A. The Chain of Amendments and Arguments During Prosecution Make Clear that M<sup>1</sup> Requires Cobalt.

During prosecution of the '035 Patent, Maxell and the examiner engaged in significant back and forth illuminating the scope of what eventually became the Cobalt Term. The original application required that for all claims, "M¹ represents at least one transition metal element

selected from Co, Ni and Mn."<sup>5</sup> Appx0214. The examiner issued an election/restriction office action, requiring Maxell to "[p]lease define M1 as x) Co or y) Ni or z) Mn [please pick one from (x)-(z)]." Appx0511 (second bracketed text and emphasis in original). In response, Maxell clearly and unequivocally elected that "in the formula Li<sub>a</sub>M¹<sub>b</sub>M²<sub>c</sub>M³<sub>d</sub>O₂ M¹ is x) CO." Appx0517 (emphases in original). This election early during prosecution informs a POSITA regarding the importance of cobalt in the material M¹ and establishes that M¹ requires cobalt.

Later claim amendments during prosecution, and Maxell's explanation of how those amendments distinguished the claims from the prior art, further reinforce that cobalt is a necessary component of M<sup>1</sup>. Specifically, on May 19, 2014, Maxell amended Claim 1, in relevant part, to read:

wherein M<sup>1</sup> represents at least one transition metal element selected from Co, Ni and Mn, . . . .

Appx1251; see Appx1259. This amendment thus specified that M<sup>1</sup> was made up **solely** of cobalt. Distinguishing a prior art reference relied upon

<sup>&</sup>lt;sup>5</sup> The language appeared in original Claim 3 filed with the application, Appx0213-0214, which was later merged into Claim 1, Appx1209; Appx1214.

by the examiner,  $Oda\ et\ al.$  (JP 2005-302507), Maxell relied on this composition of  $M^1$ , explaining that the Oda reference "does <u>not</u> describe or teach that the transition metal consists of Co alone, since [Oda] predominantly uses Ni as a transition metal." Appx1260 (emphasis in original).

Shortly thereafter, on June 19, 2014, in reply to an examiner's advisory action indicating that the prior amendments still failed to place the application in condition for allowance, Appx1267, Maxell again amended Claim 1, in relevant part, as follows:

wherein  $M^1$  represents at least one transition metal element selected from Co, Ni and Mn, . . .

. . .

wherein the content of Co in the transition metal  $M^1$  is 30% by mole or more.

Appx1272-1273. Although the new amendment allowed M¹ to also contain nickel or manganese, the addition of the content requirement for cobalt—at the same time—ensured that at least some cobalt remained required, consistent with the history of the prosecution.

Indeed, Maxell continued to distinguish the prior art based on the required presence of cobalt in the newly amended claim. Emphasizing precisely the two amended limitations quoted above and the requirement

for at least 30% cobalt, see Appx1279-1280, Maxell again distinguished the Oda reference by arguing that "[Oda] does <u>not</u> describe or teach such a content of Co, since [Oda] predominantly uses Ni as a transition metal," Appx1280 (bold emphasis added). This prosecution history clearly and unambiguously demonstrates that the "content of Co" limitation requiring at least 30% cobalt, which later became the second part of the Cobalt Term, maintains the requirement for cobalt and was a distinguishing characteristic over the cited prior art.

The examiner clearly agreed, because in the next office action, the examiner dropped all prior art-based rejections. See Appx1293-1299. In other words, the examiner understood that cobalt was a necessary component of M¹ and was able to apply that understanding in order to withdraw her prior rejection based on Oda. The examiner's ability to compare the claims to the prior art provides additional support that the Cobalt Term is not indefinite. See Sonix, 844 F.3d at 1379 (finding relevant in overturning finding of indefiniteness that examiner in reexamination had no "apparent difficulty in determining the scope" of the term and "was able to understand and apply the term in performing a search for prior art and mak[ing] an initial rejection").

It is "difficult to believe that a [POSITA] . . . who had read the . . . relevant prosecution history would be unable to determine with reasonable certainty" whether cobalt is required. *Tinnus*, 733 F. App'x at 1015. Reading Maxell's remarks specifically explaining the necessity of "such a content of Co," there can be no doubt that the Cobalt Term requires cobalt as a necessary component of M¹. In light of this clear and unambiguous guidance in the prosecution history, which the examiner understood and applied, a POSITA would have more than reasonable certainty that the Cobalt Term requires cobalt as a necessary component of M¹.

## B. The Examiner's Section 112 Rejections Reflect a Clear Understanding that Cobalt Is a Required Element in $M^1$ .

In addition to Maxell's amendments and remarks during prosecution specifically informing a POSITA that cobalt is a necessary component of M<sup>1</sup>, the prosecution history also reveals that Maxell further satisfied the examiner with respect to Section 112 rejections relating to the Cobalt Term that reflect a clear understanding that cobalt is required in M<sup>1</sup>. Following the amendments discussed above, the examiner dropped all prior art-based rejections and introduced a number of Section 112 objections, including two addressed to the presence of cobalt in M<sup>1</sup>.

Appx1295-1298. Specifically, the examiner raised the following rejections:

10. Claim[] 1 . . . [is] rejected under 35 U.S.C. 112(a) or 35 U.S.C. 112 (pre-AIA), first paragraph, as based on a disclosure which is not enabling. The upper range of the content of the Co in the transition metal M1 is critical or essential to the practice of the invention . . . .

. . .

13. Claim[] 1 . . . [is] rejected under 35 U.S.C. 112(b) or 35 U.S.C. 112 (pre-AIA), second paragraph, as being indefinite . . . .

In claim 1 it is unclear which M1, Formula (1) or Formula (2) is being discussed in the phrase "wherein the content of Co in the transition metal M1 is 30% by mole".

Appx1297-1298. Critically, the examiner did **not** issue an indefiniteness rejection based on any lack of clarity as to whether the claims require cobalt in  $M^1$ . Indeed, each of these rejections refutes the District Court's uncertainty over whether cobalt is required because they reflect the understanding that cobalt would necessarily be present in  $M^1$ . The enablement rejection clearly specifies that the "upper range of the **content of the Co** in the transition metal M1 is **critical or essential** to the practice of the invention," undeniably recognizing that some content of cobalt is necessary to  $M^1$ . Appx1297 (emphases added). Similarly, the

indefiniteness rejection was based on a lack of specificity regarding which formula required the presence of cobalt, thus presupposing the necessary presence of cobalt. Appx1298.

In response, Maxell amended Claim 1, giving the Cobalt Term its current form:

wherein  $M^1$  represents at least one transition metal element selected from Co, Ni and Mn, . . .

. . .

wherein the content of Co in the transition metal  $M^1$  of the formulae (1) and (2) is from 30% by mole or more to 100% by mole, . . . .

Appx1303-1304. The examiner accepted that these amendments resolved the Section 112 rejections and issued a notice of allowance. Appx1320, Appx1325.

Here, the examiner reviewed the claims, issued other Section 112 rejections, and was satisfied that the claims were not indefinite. See Ancora, 744 F.3d at 739 (affirming term not indefinite in part because "examiner was clearly satisfied both as to anticipation and as to indefiniteness" after amendment to overcome indefiniteness rejection (emphases in original)). The District Court improperly ignored the prosecution history in its analysis and reached the wrong result. A POSITA reviewing

this prosecution history would have no doubt that cobalt is a necessary component of  $M^1$ .

#### **CONCLUSION**

"Indefiniteness must be proven by clear and convincing evidence." *Sonix*, 844 F.3d at 1377. Here, the Cobalt Term lies far from that standard. In view of the unambiguous language of the Cobalt Term, the specification, and the prosecution history, there can be no doubt that the element cobalt is required in M¹. Thus, the Cobalt Term is not indefinite.

For the reasons provided herein, Plaintiff-Appellant Maxell, Ltd. respectfully requests that the Court (1) vacate the District Court's judgment; (2) reverse its holding that the term "M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole" in Claim 1 of U.S. Patent No. 9,077,035 is indefinite; and (3) hold that the Cobalt Term should be given its plain and ordinary meaning.

Dated: January 30, 2023 Respectfully submitted,

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**CERTIFICATE OF SERVICE** 

I hereby certify that I electronically filed the foregoing brief with

the Clerk of the Court for the United States Court of Appeals for the

Federal Circuit by using the appellate CM/ECF system on January 30,

2023.

All counsel of record in this case are registered CM/ECF users and

will be served by the appellate CM/ECF system.

Dated: January 30, 2023

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**CERTIFICATE OF COMPLIANCE** 

1. This brief complies with the type-volume limitation of Federal

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parts of the document exempted by Federal Rule of Appellate Procedure

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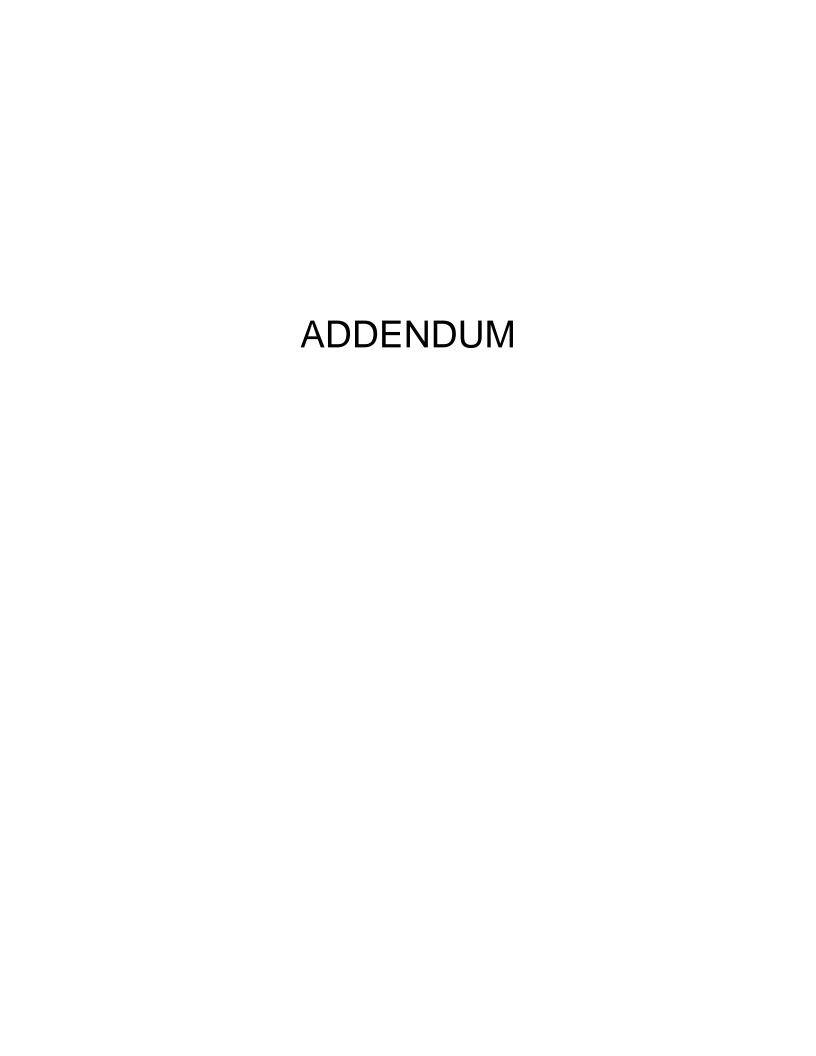
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#### IN THE UNITED STATES DISTRICT COURT FOR THE WESTERN DISTRICT OF TEXAS WACO DIVISION

MAXELL, LTD.,

Plaintiff,

AMPEREX TECHNOLOGY LIMITED,

Defendant.

AMPEREX TECHNOLOGY LIMITED,

Plaintiff,

v.

v.

MAXELL, LTD.,

Defendant.

CIVIL ACTION NO. 6:21-cv-347-ADA [LEAD CASE]

JURY TRIAL DEMANDED

CIVIL ACTION NO. 6:21-cv-1007-ADA

**JURY TRIAL DEMANDED** 

#### PARTIAL FINAL JUDGMENT PURSUANT TO RULE 54(b)

Before the Court is plaintiff Maxell, Ltd.'s ("Maxell's") Opposed Motion for Partial Reconsideration of the Court's Claim Construction Order (Dkt. No. 72), or, Alternatively, Entry of Partial Final Judgment (Dkt. No. 79, "the Motion").

Having considered the Motion, defendant Amperex Technology Limited's opposition (Dkt. No. 82), Maxell's reply (Dkt. No. 84), as well as the arguments of the parties at a hearing held on July 27, 2022, the Court declines to reconsider its claim construction order (Dkt. No. 72, the "Order") and finds that Maxell's request for partial reconsideration of the Order should be, and hereby is, **DENIED**. However, the Court further finds that Maxell's alternative request for partial final judgement should be, and hereby is **GRANTED**. Accordingly, the Court hereby severs the

U.S. Patent No. 9,077,035 (the "'035 Patent") from the balance of the case and enters partial final judgement in favor of Amperex Technology Ltd. ("ATL") and against Maxell with respect to all claims involving the '035 Patent based on the Order and the Court's supplemental claim construction opinion (ECF No. 156). The Court hereby enters the following:

- 1. The Order held that independent claim 1 of the '035 Patent is indefinite (*see* Order at 5; ECF No. 156 at 47) for the reasons set forth at oral argument and in the Court's supplemental claim construction opinion.
- 2. Each and every remaining claim of the '035 Patent depends from or incorporates independent claim 1. As such, all of the claims of the '035 Patent are indefinite for the same reason set forth in connection with claim 1.
- 3. As all claims of the '035 Patent are indefinite, the Court grants **SUMMARY JUDGMENT** that claims 1–11 of the '035 Patent are invalid.
- 4. Such holding further precludes a determination of infringement of any asserted claim of the '035 Patent, thus disposing of each of the claims and counterclaims involving the '035 Patent in the above-captioned litigations.
- 5. The Court therefore enters **FINAL JUDGMENT** in favor of defendant ATL and against Maxell with respect to Count III of Maxell's Complaint for Patent Infringement (-347 Action, Dkt. No. 1 at 39–50), Count III of ATL's Amended Complaint for Declaratory Judgment (-1007 Action, Dkt. No. 12 at 9–10), and Maxell's Third Counterclaim to ATL's Amended Complaint for Declaratory Judgment (-1007 Action, Dkt. No. 87 at 48–58) (collectively, the "'035 Patent Claims").
- 6. The Court finds that the '035 Patent Claims are distinct and separable from the remaining pending claims and counterclaims in the above-captioned litigations, and therefore the

'035 Patent Claims should be severed from the remaining pending claims and counterclaims.

- 7. The above **FINAL JUDGMENT** is "final" within the meaning of Federal Rule of Civil Procedure 54(b) because it has ended the dispute with respect to the '035 Patent on the merits, leaving nothing for the Court to do but execute the judgment. ATL is the prevailing party with respect to the '035 Patent Claims and this judgment.
  - 8. Costs incurred regarding the '035 Patent Claims are taxed against Maxell.
- 9. The Court further finds that, in the interest of sound judicial administration, there is no just reason for delaying the entry of the above **FINAL JUDGMENT**, and therefore certifies this judgment as final pursuant to Rule 54(b).

UNITED STATES DISTRICT JU

IT IS SO ORDERED.

SIGNED this 10th day of November, 2022

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## IN THE UNITED STATES DISTRICT COURT FOR THE WESTERN DISTRICT OF TEXAS WACO DIVISION

MAXELL, LTD.,

Plaintiff

-v
W-21-CV-00347-ADA

AMPEREX TECHNOLOGY LIMITED,

Defendant

Note: The content of the content of

# **CLAIM CONSTRUCTION ORDER**

The Court held a *Markman* hearing on February 14, 2022. During that hearing, the Court provided its final constructions. The Court now enters those claim constructions.

**SIGNED** this 14th day of February, 2021.

ALAN D ALBRIGHT UNITED STATES DISTRICT JUDGE

Term	Plaintiff's Proposed Construction	Defendants' Proposed Construction	Court's Preliminary Construction
#1: "positive electrode mixture layer" U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,077,035, Cl. 1;	No construction necessary; Plain and ordinary meaning	"A mixture of at least two lithium-containing transition metal oxides formed on one or both sides of an electrode current collector"	Plain-and-ordinary meaning
#2: "the positive electrode contains, as [an] active material[s], at least two lithium-containing transition metal oxides having different average particle sizes."  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,350,019, Cl. 1;	No construction necessary; Plain and ordinary meaning	"The active material of the positive electrode is a mixture formed from two or more lithium-containing transition metal oxides, at least two of the transition metal oxides having different average particle sizes"	Plain-and-ordinary meaning

Term	Plaintiff's Proposed Construction	Defendants' Proposed Construction	Court's Preliminary Construction
#3: "different compositions of elements" / "is different from" U.S. Patent No. 8,691,446 Patent, Cls. 1, 2, 3 U.S. Patent No. 9,350,019, Cls. 3, 4	No construction necessary; Plain and ordinary meaning	"The first and second lithium-containing transition metal oxides do not share all of the same chemical elements"	Plain-and-ordinary meaning
#4: "a compound having at least two nitrile groups" U.S. Patent No. 8,691,446 Patent, Cls. 1, 3 U.S. Patent No. 9,350,019, Cls. 1	No construction necessary; Plain and ordinary meaning	"A compound with at least two nitrile groups that are 1% or less by total weight of the electrolyte"	Plain-and-ordinary meaning
#5: "0.97 < x<1.02, 0.8 9.8 9.8 9.9 9.002 9.2 9.0 9.8 9.8 9.9 9.8 9.9 9.9 9.9 9.8 9.9 9.9 U.S. Patent No. 8,691,446  Patent, Cls. 1, 2, 3, 4  U.S. Patent No. 9,350,019, Cls. 1, 2; U.S. Patent No. 9,350,019, Cls. 1, 2; U.S. Patent No. 9,350,035, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and- ordinary meaning.

Term	Plaintiff's Proposed	Defendants' Proposed	Court's Preliminary
	Construction	Construction	Construction
#6: "M² represents Mg, or Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn, [and] a content of Mg in the formula (1) is from 0.15% by mole to less than 2% by mole based on an amount of the metal M¹"	ury;	Indefinite	Not indefinite. Plain-and-ordinary meaning.
U.S. Patent No. 9,350,019, Cl. 1			
#7: "at least 3.5 g/cm³".  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3 U.S. Patent No. 9,350,019, Cls. 1; U.S. Patent No. 9,077,035, Cl. 7	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and-ordinary meaning.

Term	Plaintiff's Proposed Construction	Defendants' Proposed Construction	Court's Preliminary Construction
#8: "M¹ represents at least one transition metal element selected from Co, Ni and Mn, wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole."  U.S. Patent No. 9,077,035, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite	Indefinite
#9: "the shutdown layer has a thickness A (μm) of 5 to 30, the heatresistant layer has a thickness B (μm) of 1 to 10, a sum of A and B is 6 to 23, and a ratio A/B is ½ to 4"  U.S. Patent No. 9,166,251, CI.	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and- ordinary meaning.
#10: "a particle size of 0.2 µm or less a particle size of 2 µm or more"  U.S. Patent No. 9,166,251, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and- ordinary meaning.

## IN THE UNITED STATES DISTRICT COURT FOR THE WESTERN DISTRICT OF TEXAS WACO DIVISION

MAXELL, LTD.,

Plaintiff

-v
W-21-CV-00347-ADA

AMPEREX TECHNOLOGY LIMITED,

Defendant

Note: The plaintiff of the plaintif

#### CLAIM CONSTRUCTION ORDER AND MEMORANDUM IN SUPPORT THEREOF

Before the Court are the Parties' claim construction briefs: Defendant Amperex Technology Limited's Opening and Reply briefs (ECF Nos. 51 and 54 respectively) and Plaintiff Maxell Limited's Response and Sur-Reply briefs (ECF Nos. 52 and 58, respectively). The Court provided preliminary constructions for the disputed terms one day before the hearing. The Court held the *Markman* hearing on February 14, 2022. ECF No. 69. During that hearing, the Court informed the Parties of the final constructions for the disputed terms. *Id.* This Order does not alter any of those constructions.

#### I. DESCRIPTION OF THE ASSERTED PATENTS

Plaintiff asserts U.S. Patent Nos. 8,691,446, 9,077,035, 9,166,251, and 9,350,019. The '446, '035, and '019 Patents are directed towards the chemical composition of a battery's positive electrode, while the '251 Patent is directed towards separating battery components using heat-resistant elements.

#### A. The '446, '035, and '019 Patents

The '446, '035, and '019 Patents are all entitled "Nonaqueous secondary battery and method of using the same." The '446 Patent is the parent of the '019 Patent; the '035 Patent is unrelated to the '446 and '035 Patents, but appears to be directed to similar technology and shares many of the same claim terms. *See, e.g.*, '446 Patent, Claim 1; '035 Patent, Claim 1; '019 Patent, Claim 1; *see also* Opening at 3–4.

Each of these patents describes a lithium-ion battery whose positive electrode includes at least two lithium-containing transition metal oxides with different average particle sizes. *See, e.g.*, '446 Patent, Claim 1, Limitation [c] ("wherein the positive electrode contains, as an active material, at least two lithium containing transition metal oxides having different average particle sizes"); '035 Patent, Claim 1, Limitation [b] (same); '019 Patent, Claim 1, Limitation [b] (same). Using two lithium-containing transition metal oxides with different average particle sizes allows the battery manufacturer to increase the number of lithium-containing transition metal oxides in the same volume, which allows for a more compact battery, as well as greater energy efficiency.

#### B. The '251 Patent

The '251 Patent is entitled "battery separator and nonaqueous electrolyte battery." The '251 Patent purports to disclose a novel separator between the positive and negative portions of the lithium-ion battery. '251 Patent at Abstract. More specifically, the '251 Patent recites:

In the nonaqueous electrolyte battery of the present invention, the heat generation starting temperature of the positive electrode is as high as 180° C. or higher. This can suppress heat to be generated from the positive electrode when the battery is placed in a high-temperature environment. Moreover, the nonaqueous electrolyte battery uses the battery separator that includes a thermoplastic resin and heat-

resistant fine particles containing particles with a particle size of  $0.2~\mu m$  or less in a proportion of 10~vol% or less and particles with a particle size of  $2~\mu m$  or more in a proportion of 10~vol% or less, and that effects a shutdown in the range of  $100^\circ$  C. to  $150^\circ$  C. This can reliably prevent a short circuit due to contact between the positive electrode and the negative electrode at high temperatures.

*Id.* at 2:43–56.

#### II. LEGAL STANDARD

#### A. General principles

The general rule is that claim terms are generally given their plain-and-ordinary meaning. *Phillips v. AWH Corp.*, 415 F.3d 1303, 1312 (Fed. Cir. 2005) (*en banc*); *Azure Networks, LLC v. CSR PLC*, 771 F.3d 1336, 1347 (Fed. Cir. 2014), *vacated on other grounds*, 575 U.S. 959, 959 (2015) ("There is a heavy presumption that claim terms carry their accustomed meaning in the relevant com3333munity at the relevant time.") (internal quotation omitted). The plain-and-ordinary meaning of a term is the "meaning that the term would have to a person of ordinary skill in the art in question at the time of the invention." *Phillips*, 415 F.3d at 1313.

The "only two exceptions to [the] general rule" that claim terms are construed according to their plain-and-ordinary meaning are when the patentee (1) acts as his/her own lexicographer or (2) disavows the full scope of the claim term either in the specification or during prosecution. *Thorner v. Sony Computer Ent. Am. LLC*, 669 F.3d 1362, 1365 (Fed. Cir. 2012). The Federal Circuit has counseled that "[t]he standards for finding lexicography and disavowal are exacting." *Hill-Rom Servs., Inc. v. Stryker Corp.*, 755 F.3d 1367, 1371 (Fed. Cir. 2014). To act as his/her

own lexicographer, the patentee must "clearly set forth a definition of the disputed claim term" and "clearly express an intent' to [define] the term." *Thorner*, 669 F.3d at 1365.

"Like the specification, the prosecution history provides evidence of how the PTO and the inventor understood the patent." *Phillips*, 415 F.3d at 1317. "[D]istinguishing the claimed invention over the prior art, an applicant is indicating what a claim does not cover." *Spectrum Int'l, Inc. v. Sterilite Corp.*, 164 F.3d 1372, 1379 (Fed. Cir. 1998). The doctrine of prosecution disclaimer precludes a patentee from recapturing a specific meaning that was previously disclaimed during prosecution. *Omega Eng'g, Inc. v. Raytek Corp.*, 334 F.3d 1314, 1323 (Fed. Cir. 2003). "[F]or prosecution disclaimer to attach, our precedent requires that the alleged disavowing actions or statements made during prosecution be both clear and unmistakable." *Id.* at 1325–26. Accordingly, when "an applicant's statements are amenable to multiple reasonable interpretations, they cannot be deemed clear and unmistakable." *3M Innovative Props. Co. v. Tredegar Corp.*, 725 F.3d 1315, 1326 (Fed. Cir. 2013).

A construction of "plain and ordinary meaning" may be inadequate when a term has more than one "ordinary" meaning or when reliance on a term's "ordinary" meaning does not resolve the parties' dispute. *O2 Micro Int'l Ltd. v. Beyond Innovation Tech. Co.*, 521 F.3d 1351, 1361 (Fed. Cir. 2008). In that case, the Court must describe what the plain-and-ordinary meaning is. *Id.* 

"Although the specification may aid the court in interpreting the meaning of disputed claim language . . ., particular embodiments and examples appearing in the specification will not generally be read into the claims." *Constant v. Advanced Micro-Devices, Inc.*, 848 F.2d 1560, 1571

(Fed. Cir. 1988). "[I]t is improper to read limitations from a preferred embodiment described in the specification—even if it is the only embodiment—into the claims absent a clear indication in the intrinsic record that the patentee intended the claims to be so limited." *Liebel-Flarsheim Co.* v. Medrad, Inc., 358 F.3d 898, 913 (Fed. Cir. 2004).

An applicant's statements during the PCT prosecution may also indicate the scope of the invention. *See Caterpillar Tractor Co. v. Berco*, 714 F.2d 1110, 1116 (Fed. Cir. 1983) (stating that when instructions to foreign counsel or representations to foreign patent offices made by an applicant during prosecution of a corresponding foreign application provide "relevant evidence" with respect to claim interpretation, such information "must be considered."); *see also Gillette Co. v. Energizer Holdings, Inc.*, 405 F.3d 1367, 1374 (Fed. Cir. 2005) (finding that the applicant's own statements made before the European Patent office support the court's holding).

Although extrinsic evidence can be useful, it is "less significant than the intrinsic record in determining 'the legally operative meaning of claim language." *Phillips*, 415 F.3d at 1317 (quoting *C.R. Bard, Inc. v. United States Surgical Corp.*, 388 F.3d 858, 862 (Fed. Cir. 2004)). Technical dictionaries may be helpful, but they may also provide definitions that are too broad or not indicative of how the term is used in the patent. *Id.* at 1318. Expert testimony may also be helpful, but an expert's conclusory or unsupported assertions as to the meaning of a term are not. *Id.* 

## **B.** Indefiniteness

"[I]ndefiniteness is a question of law and in effect part of claim construction." *ePlus, Inc. v. Lawson Software, Inc.*, 700 F.3d 509, 517 (Fed. Cir. 2012). Patent claims must particularly point out and distinctly claim the subject matter regarded as the invention. 35 U.S.C. § 112, ¶ 2. A claim, when viewed in light of the intrinsic evidence, must "inform those skilled in the art about the scope of the invention with reasonable certainty." *Nautilus Inc. v. Biosig Instruments, Inc.*, 572 U.S. 898, 910 (2014). If it does not, the claim fails § 112, ¶ 2 and is therefore invalid as indefinite. *Id.* at 901. Whether a claim is indefinite is determined from the perspective of one of ordinary skill in the art as of the time the application was filed. *Id.* at 911.

#### III. LEGAL ANALYSIS

A. Term #1: "positive electrode mixture layer"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"positive electrode mixture layer"  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,077,035, Cl. 1	No construction necessary; Plain and ordinary meaning	"A mixture of at least two lithium-containing transition metal oxides formed on one or both sides of an electrode current collector"

The parties dispute whether this term has a plain-and-ordinary meaning and what the proper construction should be.

# 1. Whether "positive electrode mixture layer" has a plain-and-ordinary meaning

#### **The Parties' Positions:**

With respect to this dispute, Defendant contends that this term does not have a plain-and-ordinary meaning to a POSITA. Opening at 7. Rather, at least according to Defendant, this term is specific to the '446, '019, and '035 Patents. *Id*.

In its response, Plaintiff contends that this term is "well-understood" in light of the intrinsic evidence. Response at 3 (citing ECF No. 52-1 (Lucht Declaration) at ¶ 17).

In its reply, Defendant's expert opines that this term does not have a plain-and-ordinary meaning to a POSITA. Reply at 2 (citing ECF No. 51-15 (Fuller Declaration) at ¶ 25). Defendant contends that Plaintiff and Plaintiff's expert "cite no evidence whatsoever showing use of this term in the relevant literature because that term is not a term of art in the industry." *Id.* Defendant contends that "[w]hile both experts understand what the phrase 'positive electrode layer' means, [Defendant's expert] states—and [Plaintiff's expert] does not deny—that the added word 'mixture' is uncommon and thus needs an explanation informed by the specification." *Id.* Defendant contends that "[f]aced with competing experts, the Court cannot conclude without more that the disputed claims can be given their plain and ordinary meaning." *Id.* (citing *Eon Corp. IP Holdings LLC*, 815 F.3d 1314, 1319 (Fed. Cir. 2016)).

In its sur-reply, Plaintiff contends that a POSITA would understand the meaning of both "positive electrode layer" and "mixture" so a POSITA would understand the combination of the two. Sur-Reply at 2 (citing Sur-Reply, Ex. 19 (Fuller Deposition) at 36:15–19).

#### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff and that the term "positive electrode mixture layer" has a plain-and-ordinary meaning for the reasons that follow. *First*, the claims describe that the "positive electrode" comprises a "positive electrode mixture layer" and that the "positive electrode" comprises a mixture of "at least two lithium containing transition metal oxides having different average particle sizes." *See*, *e.g.*, '446 Patent, Claim 1, Lims. [a], [b]. *Second*, descriptions in the specification confirm that the term is simply the sum of its constituent words, *i.e.*, a mixture layer for the positive electrode. *See*, *e.g.*, '446 Patent at 2:53–62.

#### 2. Dispute what the proper construction should be

#### **The Parties' Positions:**

Defendant contends that the specification makes it clear that "two lithium-containing transition metal oxides are blended together in a particular weight ratio and applied to the current collector for the positive electrode[.]" Opening at 8 (citing/quoting '446 Patent at 16:62–17:1, '019 Patent at 16:54–17:9, '035 Patent at 11:65–12:3). Defendant also contends that the specification describes that this mixture is "essential." *Id.* (citing '446 Patent at 17:45–47, '019 Patent at 17:53–55, '035 Patent at 13:20–22). Defendant contends that the "mixture must include at least two different lithium-containing transition metal oxides." *Id.* (quoting '446 Patent at 9:51–62, '019 Patent at 9:59–10:3, '035 Patent at 4:66-5:1–10).

Defendant contends that the specification makes it clear that the mixture layer is "formed on" the surfaces of the electrode collector. *Id.* (citing '446 Patent at 7:53–57, '019 Patent at 7:61–65). Defendant contends that the specification also makes it clear that the mixture is "uniformly applied to both surfaces" of the collector. *Id.* (citing '035 Patent at 22:9–13). Finally, Defendant contends that the specifications provide examples of the "present invention," where each example describes that the "positive electrode mixture layer" includes "two or more transition metal oxides blended together and applied to both surfaces of the electrode collector." *Id.* at 8–9 (citing '446 Patent at 22:30–27:23, '019 Patent at 22:39–27:29, '035 Patent at 21:34–26:49).

In its response, Plaintiff contends that Defendant's proposed construction excludes known, necessary components from the electrode layer. Response at 3. More specifically, Plaintiff contends that Defendant's proposed construction "improperly omits the presence of additional materials (*e.g.*, a binder or an electric conductive aid) likely to be present in the positive electrode mixture layer." *Id.* Plaintiff contends that a POSITA would understand that a binder is often a necessary part of the layer. *Id.* at 4 (citing ECF No. 52-1 (Lucht Declaration) at ¶¶ 35–37). Plaintiff contends that the specification discloses the presence of the binder in the positive electrode mixture layer. *Id.* at 3–4 (quoting/citing '446 Patent at 16:47–16:57, 17:14–19; '019 Patent at 16:55–17:12, 17:22-24; '035 Patent at 11:66-12:18, 12:56-58).

Plaintiff next contends that Defendant's proposed construction improperly attempts to add an unsupported process limitation. *Id.* at 4. More specifically, Plaintiff contends that the specifications of the '446, '035, and '019 Patents "do not require that the positive electrode active material be applied to the electrode current collector using any particular deposition method (*e.g.*,

'formed on')." *Id.* Plaintiff contends that it is inappropriate to add a process limitation to an apparatus claim, unless the "process is an essential feature of the apparatus." *Id.* (citing *Medtronic Vascular Inc. v. Bos. Sci. Corp.*, 526 F. Supp. 2d 613, 622–23 (E.D. Tex. 2007)). Plaintiff contends that "formed on" is not essential. *Id.* (citing ECF No. 52-1 (Lucht Declaration) at ¶ 38, '446 Patent at 17:2–4 ("the method for producing the positive electrode is not limited . . . and may be any other method.")).

In its reply, with respect to Plaintiff's argument that Defendant's proposed construction excludes a binder or an electric conductive aid, Defendant contends that the "at least" language in its proposed construction does not exclude anything, but rather only specifies what is needed. Reply at 2. With respect to whether Defendant's proposed construction improperly adds a process limitation, Defendant contends that "mixture layer' already includes an embedded process," as every embodiment in in the patents begins with mixing metals together. *Id.* at 2–3.

In its sur-reply, with respect to Plaintiff's argument that Defendant's proposed construction excludes a binder or an electric conductive aid, Plaintiff contends that "at least" modifies "two" which limits the mixture to lithium-containing transition metal oxides and nothing else. Sur-Reply at 3. With respect to whether Defendant's proposed construction improperly adds a process limitation, Plaintiff contends that "formed on" improperly imports a limitation from the specification.

#### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff and finds that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347. *Second*, Defendant does not expressly allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning. *Thorner*, 669 F.3d at 1365.

Third, part of Defendant's proposed construction renders some claim language to be superfluous. For example, Claim 1, Limitation [b] of the '446 Patent provides "wherein the positive electrode contains, as an active material, at least two lithium containing transition metal oxides having different average particle sizes[.]" See also '019 Patent, Claim 1, Limitation [b] (same); '035 Patent, Claim 1, Limitation [b] (same). As such, Defendant's proposed construction use of the phrase "mixture of at least two lithium-containing transmission metal oxides" renders most of those limitations to be superfluous. Generation II Orthotics Inc. v. Med. Tech. Inc., 263 F.3d 1356, 1365 (Fed. Cir. 2001) (a construction should not "revise or ignore the explicit language of the claims.").

Fourth, the Court finds that Defendant's proposed construction use of the phrase "formed on" improperly imports a limitation from the specification. *Phillips*, 415 F.3d at 1320 ("One of the cardinal sins of patent law [is] reading a limitation from the written description into the

claims.") (quoting SciMed Life Sys., Inc. v. Advanced Cardiovascular Sys., Inc., 242 F.3d 1337, 1340 (Fed. Cir. 2001).

Fifth, the Court disagrees with Defendant that the "present invention" language in the specification limits the scope of this claim term. Defendant cites '446 Patent at 22:30–27:23, '019 Patent at 22:39–27:29, '035 Patent at 21:34–26:49 in support of its argument that the specifications describe the "present invention" using examples and that those examples describe that the "positive electrode mixture layer" includes "two or more transition metal oxides blended together and applied to both surfaces of the electrode collector." Opening at 8–9. But the specifications of all three patents expressly describe that "the Examples do not limit the present invention." '446 Patent at 22:33-34, '019 Patent at 22:42-43, '035 Patent at 21:37-38. Given that the specifications expressly recite that the present invention is not limited to the disclosed examples and Defendant has not provided a reason why the Court should ignore that express language and/or why the present invention is limited to those examples, the Court declines to limit the "positive electrode mixture layer" to require "two or more transition metal oxides blended together and applied to both surfaces of the electrode collector." Rambus, Inc. v. Infineon Techs. AG, 318 F.3d 1081, 1094–95 (Fed. Cir. 2003) (although portions of the written description referred to the term at issue as limiting the claimed invention to a preferred embodiment, "the remainder of the specification and the prosecution history shows that Rambus did not clearly disclaim or disavow such claim scope in this case.").

On the other hand, the Court disagrees with Plaintiff that the "at least two lithium-containing transition metal oxides" excludes other structures, e.g., a binder or an electric

conductive aid. Rather, that language appears only to specify the minimum number of lithium-containing transition metal oxides that the claimed invention requires.

Therefore, for the reasons described above, the Court's final construction for "positive electrode mixture layer" is plain-and-ordinary meaning.

B. Term #2: "the positive electrode contains, as [an] active material[s], at least two lithium-containing transition metal oxides having different average particle sizes"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"the positive electrode contains, as [an] active material[s], at least two lithium-containing transition metal oxides having different average particle sizes"  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,077,035, Cl. 1	No construction necessary; Plain and ordinary meaning	"The active material of the positive electrode is a mixture formed from two or more lithium-containing transition metal oxides, at least two of the transition metal oxides having different average particle sizes"

## **The Parties' Positions:**

Defendant contends that its proposed construction is the plain-and-ordinary meaning. Opening at 9. Defendant contends that the specification only describes <u>forming</u> the positive electrode in one way with "at least two lithium-containing transition metals with different average particle sizes." *Id.* at 10 (quoting/citing '446 Patent at 16:46–66, '019 Patent at 16:54–17:7, '035 Patent at 11:65-12:15). Defendant contends that the specification describes "formation of the positive electrode by a specific mixture as 'essential." *Id.* at 11 (quoting/citing '446 Patent at

17:45–47, '019 Patent at 17:53–55, '035 Patent at 13:20–23; Sunrace Roots Enter. Co., Ltd. v. SRAM Corp., 336 F.3d 1298, 1305 (Fed. Cir. 2003)).

In its response, Plaintiff contends that Defendant's proposed construction improperly limits the "only" active material to a mixture of two or more lithium-containing transition metal oxides. Response at 5. Plaintiff contends that, by contrast, a POSITA would understand that there can be other active materials. *Id.* at 5–6.

Plaintiff also contends that Defendant is attempting—as it did for Term #1—to read a process limitation into the claims. *Id.* at 6. But Plaintiff contends that the only time that this is permissible is when the specification describes that that process as "essential." *Id.* Plaintiff contends that the specification focuses on the composition of the positive electrode, and not a process of creating it. *Id.* 

In its reply, with respect to Plaintiff's argument that Defendant's proposed construction limits what the "active material" may be, Defendant contends that the "at least" language in its proposed construction does not exclude anything, but rather only specifies what is needed. Reply at 3. With respect to the "formed on" language in its proposed construction, Defendant contends that "[b]ecause the patents each describe the formation of the positive electrode by a specific mixture as 'essential,' the construction should be limited to what 'was described as essential to the invention." *Id.* at 4 (citing '446 Patent at 17:45-47, '019 Patent at 17:53-55, '035 Patent at 13:20-23; *Sunrace*, 336 F.3d at 1305).

In its sur-reply, Plaintiff repeats its contention that Defendant's proposed construction improperly limits the "only" active material to a mixture of two or more lithium-containing

transition metal oxides. Sur-Reply at 4. Plaintiff contends that Defendant "does not explain in its reply brief how its proposed construction could include materials other than lithium-containing transition metal oxides." *Id*.

Plaintiff also repeats its contention that Defendant is improperly attempting to read a process limitation into the claims. *Id.* at 4. Plaintiff contends even Defendant's expert admits that "formed on" does not appear in the claims. *Id.* (quoting Sur-Reply, Ex. 19 (Fuller Deposition) at 59:16–19).

#### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff and finds that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347. *Second*, Defendant does not expressly allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning. *Thorner*, 669 F.3d at 1365.

*Third*, the Court finds that Defendant's proposed construction use of the phrase "formed on" improperly imports a limitation from the specification. *Phillips*, 415 F.3d at 1320 ("One of the cardinal sins of patent law [is] reading a limitation from the written description into the claims.") (quoting *SciMed Life*, 242 F.3d at 1340).

Fourth, the Court disagrees with Defendant that the specification describes "formation of the positive electrode by a specific mixture as 'essential." Response at 11. The passages that Defendant cites in support of that conclusion recites that "[i]t is essential for the nonaqueous secondary battery of the present invention to have the nonaqueous electrolyte and the positive electrode, which are explained above, and thus there is no specific limitation on other elements or structure of the battery." '446 Patent at 17:45–47, '019 Patent at 17:53–55, '035 Patent at 13:20–23. This passage only generally describes that it is "essential" that the "nonaqueous secondary battery of the present invention to have the nonaqueous electrolyte and the positive electrode." It does not, however, specifically describe that the formation of the positive electrode by a specific mixture as "essential."

Fifth, apart from the "formed from" wording, Defendant's proposed construction appears to otherwise simply paraphrase the claim language, which the Court declines to do. C.R. Bard, Inc. v. United States Surgical Corp., 388 F.3d 858, 863 (Fed. Cir. 2004) (agreeing that "merely rephrasing or paraphrasing the plain language of a claim by substituting synonyms does not represent genuine claim construction") (internal quotations omitted).

On the other hand, the Court disagrees with Plaintiff that the "at least two lithium-containing transition metal oxides" excludes other structures, *e.g.*, a binder or an electric conductive aid. Rather, that language appears only to specify the minimum number of lithium-containing transition metal oxides that the claimed invention requires.

Therefore, for the reasons described above, the Court's final construction for this term is plain-and-ordinary meaning.

C. Term #3: "different compositions of elements" / "is different from"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"different compositions of elements" / "is different from"  U.S. Patent No. 8,691,446 Patent, Cls. 1, 2, 3 U.S. Patent No. 9,350,019, Cls. 3, 4	No construction necessary; Plain and ordinary meaning	"The first and second lithium- containing transition metal oxides do not share all of the same chemical elements"

#### **The Parties' Positions:**

The dispute between the parties is whether the elements need to be different or whether the elements can be the same (or different) but have different molecular proportions.

Defendant contends that the specifications describe that a "different composition" of elements have different elements. Opening at 13 (citing '446 Patent at 15:7–17, '019 Patent at 15:15–25; *see also* '446 Patent at 27:11–23, '019 Patent at 27:16–29). On the other hand, Defendants contend that while the specifications disclose molecules with the same elements, but different proportions, are not "different compositions of elements," but are rather the "same composition of elements." *Id.* at 14. Defendant contends that Examples 9 and 12 "illustrate" different composition of elements, namely that "[E]xample 9 depicts metal oxide A as containing LiCo and metal oxide B as containing LiCu, while Example 12 depicts metal oxide A as containing LiCu versus metal oxide B containing LiCo plus Ni and Mn." *Id.* (citing '446 Patent at 26:30–44, 27:11–23; '019 Patent at 26:37–51, 27:16–29). Defendant contends that Examples 1–8 and 10-11, on the other hand, all describe metal oxides containing the same composition of elements, but

with varying molar amounts. *Id.* Defendant contends that these examples are unclaimed, and thus dedicated to the public. *Id.* at 15.

Defendant contends that during prosecution, Examiner rejected the claims in view of the Kikuchi prior art reference which disclosed "multiple lithium transition metal oxide compounds with different particular sizes." *Id.* at 14 (citing Opening, Ex. 7 at 2–3, 15). Defendant contends that Applicant amended the claims to add "different compositions." *Id.* at 14–15 (citing Opening, Ex. 8 at 2, 4, 5, 6). Defendant contends that Applicant argued that, unlike the claimed invention, Kikuchi disclosed the "same composition of elements." *Id.* at 15 (citing Opening, Ex. 8 at 13). Defendant contends that the claims of the '446 and '019 Patents were only allowed over Kikuchi because the claims were amended to claim two chemically different lithium-containing transition metal oxides. *Id.* 

In its response, Plaintiff contends that the terms do not require construction as their meaning is "ascertainable in light of the intrinsic evidence." Response at 6. Plaintiff contends that the term "clearly encompasses two scenarios: (1) the first and second lithium-containing transition metal oxides do not share all of the same chemical elements; and (2) the first and second lithium-containing transition metal oxides contain all of the same chemical elements, but in different proportions." *Id.* at 6–7.

Plaintiff also contend that certain claims require "different" compositions while using the same variables for specific elements, *e.g.*, Li<sub>a</sub>M<sup>1</sup><sub>b</sub>M<sup>2</sup><sub>c</sub>M<sup>3</sup><sub>d</sub>O<sub>2</sub> and Li<sub>x</sub>M<sup>1</sup><sub>y</sub>M<sup>2</sup><sub>z</sub>M<sup>3</sup><sub>v</sub>O<sub>2</sub>, where M<sup>#</sup> are the different elements. Response at 7 (citing '446 Patent, Claims 1–3, '019 Patent, Claims 3–4). Plaintiff contends that because the elements M<sup>#</sup> are the same between the two compositions, a

POSITA would understand that the "two formulas could differ either in the selection of elements for M<sup>1</sup>, M<sup>2</sup>, and M<sup>3</sup>, or in the selection of the elemental ratios." *Id.* Based on that, Plaintiff contends Defendant's proposed construction (which excludes compositions that use the same elements) is contrary to the claim language. *Id.* 

Plaintiff contends that Defendant's proposed construction is also contrary to the specification. *Id.* In particular, Plaintiff contends that the specification recites "M¹, M², and M³ are selected from the same elements as in the formula (1), **but the elements selected or the constituting element ratios selected** in the individual positive electrode active materials having different average particle sizes **may differ from each other**." *Id.* at 7–8 (quoting '446 Patent at 11:21–25 (emphasis in Plaintiff's brief)).

Plaintiff further contends that Defendant's proposed construction departs from the basic principles of chemistry. *Id.* at 8. Basic chemistry dictates that different ratios results in different compounds, *e.g.*, H<sub>2</sub>O versus H<sub>2</sub>O<sub>2</sub>. *Id*.

Plaintiff contends that Defendants ignore numerous disclosures in the specification in order to limit the claim to a single embodiment. *Id.* Plaintiff contends that only one of the examples provided in the '446, '019, and '035 Patents "describes an embodiment where the two transition lithium-containing transition metal oxides contain different chemical elements. All but one[] of the remaining examples disclose lithium-containing transition metal oxides that contain all of the same chemical elements in different proportions[.]" *Id.* at 9–10. Plaintiff contends that Defendant's proposed construction improperly excludes all of these examples. *Id.* at 10.

With respect to Defendant's prosecution history argument, Plaintiff contends that the alleged disclaimer is not "clear and unambiguous." *Id.* Plaintiff then contends that Defendant is also wrong about the prosecution history. *Id.* More specifically, Plaintiff contends that during prosecution, Applicant distinguished the amended claim by arguing:

Kikuchi JP '582 describes the primary and secondary particles, but these particles are made of the same material and should have the same composition of elements, Kikuchi JP '582 does not describe nor teach the use of lithium-containing transition metal oxides having different average particle sizes which have different compositions of elements between them.

*Id.* (citing Opening, Ex. 8 at 13.). Plaintiff contend that Kikuchi describes an invention where the large and small particles are made from the same composition of elements, *i.e.*, contain identical elements and identical subscripts for each element. *Id.* at 11–12.

In its reply, Defendant contends that Plaintiff invites the Court to construe "different" as "the same." Reply at 4. Defendant contends that if the inventor intended that "different compositions of elements" meant "the same elements but in different proportions," the inventor would have drafted the claims accordingly. *Id.* Defendant again contends that the claims do not need to cover all examples. *Id.* 

Defendant contends that during prosecution, Applicant "overcame this rejection by calling for an anode with at least two different lithium-containing transition metal oxides 'having different average particle sizes' and 'different compositions of elements between them." *Id.* (quoting Opening, Ex. 8 at 2, 4, 5, 6). Defendant contends that Applicant contrasted Kikuchi, as having the "same material and should have the same composition of elements." *Id.* (quoting Opening, Ex. 8

at 2, 4, 5, 6). Based on those statements, Defendant contends that Plaintiff cannot recapture claim scope it gave up during prosecution. *Id*.

In its sur-reply, Plaintiff again contends that the specification recites "M¹, M², and M³ are selected from the same elements as in the formula (1), but the elements selected or the constituting element ratios selected in the individual positive electrode active materials having different average particle sizes may differ from each other." Sur-Reply at 5 (quoting '446 Patent at 11:21–25). Plaintiff also contends that Defendant's expert admits that "composition" refers to the ratio of elements in a formula. *Id.* (Sur-Reply, Ex. 19 at 64:1–4). Plaintiff contends that Defendant and its expert essentially argue that "compounds with different ratios of elements are the same compound." *Id.* at 6.

Plaintiff contends that Applicant argued that the Kikuchi reference described "primary and secondary particles, but these particles are **made of the same material and should have the same composition of elements**, Kikuchi JP '582 does not describe nor teach the use of lithium-containing transition metal oxides having different average particle sizes **which have different compositions of elements between them.**" *Id.* (citing Opening, Ex. 8 at 13) (emphasis in Plaintiff's brief).

#### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff and finds that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, the "heavy presumption" is that terms should be construed according to their

plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347. *Second*, Defendant does not expressly allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning. *Thorner*, 669 F.3d at 1365.

*Third*, the claims describe that a "different composition" includes two compounds that use the same elements, but with different molar amounts. For example, Claim 1, Limitation [b] of the '446 Patent recites that the claim requires two lithium-containing transition metal oxides with "different compositions of elements." Claim 1, Limitation [c] describes that the lithium-containing transition metal oxide with the smaller particle size is governed by formula (1), which is Li<sub>x</sub>M<sup>1</sup> <sub>v</sub>M<sup>2</sup><sub>z</sub>M<sup>3</sup><sub>v</sub>O<sub>2</sub>. Dependent Claim 2 of the '446 Patent recites that the lithium-containing transition metal oxide with the larger particle size is governed by formula (2), which is Li<sub>a</sub>M<sup>1</sup><sub>b</sub>M<sup>2</sup><sub>c</sub>M<sup>3</sup><sub>d</sub>O<sub>2</sub>. Claims 1 and 2 both recite that "M1 represents at least one transition metal element selected from Co, Ni and Mn." '446 Patent, Claim 1, Limitation [d]; Claim 2, Limitation [b]. Claim 1 recites that "M<sup>2</sup> represents Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn," while Claim 2 recites that "M<sup>2</sup> represents at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn. Id., Claim 1, Limitation [d]; Claim 2, Limitation [b]. Claims 1 and 2 both recite that "M<sup>3</sup> represents an element other than Li." Id., Claim 1, Limitation [d]; Claim 2, Limitation [b]. In other words, Claims 1 and 2 recite the same requirements for M<sup>1</sup> and M<sup>3</sup>, and the requirements for M<sup>2</sup> could be the same as well. Therefore, Claims 1 and 2 of the '446 Patent describes that two compounds that could use the same elements—only in different molar amounts—are "different compositions." In the same manner, Claims 1 to 4 of the '019 Patent also describe the same relationship.

Fourth, the specifications describe that a "different composition" includes two compounds that use the same elements, but with different molar amounts. For example, the '446 Patent specification recites "M¹, M², and M³ are selected from the same elements as in the formula (1), but the elements selected or the constituting element ratios selected in the individual positive electrode active materials having different average particle sizes may differ from each other." '446 Patent at 11:21–25 (emphasis added). In other words, this passage describes that "different compositions" could cover different "constituting element ratios," while the selected elements could be the same.

*Fifth*, the Court agrees with Plaintiff that basic chemistry dictates that different ratios results in different compounds, *e.g.*, H<sub>2</sub>O versus H<sub>2</sub>O<sub>2</sub>, which may have very different properties.

**Sixth**, the Court agrees with Plaintiff that Defendant's proposed construction excludes disclosed embodiments, *e.g.*, Examples 1–8 and 10–11. *Oatey Co. v. IPS Corp.*, 514 F.3d 1271, 1276 (Fed. Cir. 2008) ("We normally do not interpret claim terms in a way that excludes embodiments disclosed in the specification. . . . where claims can reasonably be interpreted to include a specific embodiment, it is incorrect to construe the claims to exclude that embodiment, absent probative evidence to the contrary."). Defendant does not appear to provide probative evidence to the contrary, but rather asserts—without evidence or support—that the patentee intended to dedicate these examples to the public. Because unsupported attorney argument does

not rise to the level of probative evidence, the Court finds that Defendant's proposed construction improperly excludes disclosed embodiments.

Seventh, the Court agrees with Plaintiff that Defendant's proposed construction improperly limits the scope of the claim term to specific disclosed embodiments, e.g., Examples 9 and 12. Liebel-Flarsheim, 358 F.3d at 913 ("[I]t is improper to read limitations from a preferred embodiment described in the specification—even if it is the only embodiment—into the claims absent a clear indication in the intrinsic record that the patentee intended the claims to be so limited.").

Eighth, the Court agrees with Plaintiff that Applicant's prosecution statements are not "clear and unmistakable." *Id.* at 900. By contrast, the Court finds that Applicant argued that the Kikuchi reference describes a single composition ("same material" and the "same composition of elements") while the claimed invention discloses having "different compositions," *i.e.*, different material (elements) or different composition of elements. Opening, Ex. 8 at 13. At minimum, this statement equally supports that the (1) the first and second lithium-containing transition metal oxides do not share all of the same chemical elements; and (2) the first and second lithium-containing transition metal oxides contain all of the same chemical elements, but in different proportions, which indicates there is no disclaimer. *3M Innovative Props. Co. v. Tredegar Corp.*, 725 F.3d 1315, 1326 (Fed. Cir. 2013) (when "an applicant's statements are amenable to multiple reasonable interpretations, they cannot be deemed clear and unmistakable.").

Therefore, for the reasons described above, the Court's final construction for these terms is plain-and-ordinary meaning.

## D. Term #4: "a compound having at least two nitrile groups"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"a compound having at least two nitrile groups"  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cls. 1	No construction necessary; Plain and ordinary meaning	"A compound with at least two nitrile groups that are 1% or less by total weight of the electrolyte"

#### **The Parties' Positions:**

Defendant contends that the only thing the patent says about nitrile compounds is that the range is not "arbitrary," but rather the only embodiment discloses that the range is 1% or less in weight of the electrolyte solution. Opening at 16 ('446 Patent at 5:8-20, '019 Patent at 5:17-29). The reason for this, at least according to Defendant, is that the charge-discharge cycle characteristics of the battery will be degraded when the amount of the nitrile compound is too large. *Id*.

In its response, Plaintiff contends that there are two "fatal" flaws with Defendant's proposed construction. First, Plaintiff contends that Defendants' proposed construction eliminates the requirement of "two nitrile groups." Response at 12–13. Second, Plaintiff contends that it also "imports embodiments." *Id.* at 13. More specifically, Plaintiff contends that Defendant's proposed construction is based on the only disclosed embodiment, but that "it is improper to read limitations from a preferred embodiment described in the specification—even if it is the only embodiment—into the claims absent a clear indication in the intrinsic record that the patentee intended the claims to be so limited." *Id.* (quoting *Dealertrack, Inc. v. Huber*, 674 F.3d 1315, 1327 (Fed. Cir. 2012)).

In its reply, with respect to Plaintiff's second point, Defendant contends that the objective of the present invention is to provide a battery with "good charge-discharge characteristics" and that 1% or less nitrile compound is necessary to achieve that. Reply at 6. Defendant contends that "[b]ecause the patent expressly teaches away from higher concentrations of nitrile groups as 'an object of the present invention,'" the Court should adopt Defendant's proposed construction. *Id*.

In its sur-reply, Plaintiff contends that Defendant's proposed construction "drastically departs from the plain meaning of the term by incorrectly omitting the required presence of two nitrile groups and importing a numerical limitation[.]" Sur-Reply at 7. With respect to the first "fatal" flaw, Plaintiff contends that Defendant and its expert take opposite positions on whether Defendant's proposed construction requires two nitrile groups. *Id*.

With respect to the second "fatal" flaw, Plaintiff again contends that Defendant's proposed construction improperly imports an embodiment. *Id.* at 7–8.

#### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff and finds that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347.

**Second**, Defendant does not expressly allege lexicography, which is one exception to the general rule that a term should be construed as having its plain-and-ordinary meaning. *Thorner*, 669 F.3d at 1365.

Third, the Court disagrees with Defendant that the "present invention" language constitutes a disclaimer. The "present invention" passages that Defendant cite do not restrict the nitrile weight to a particular range. '446 Patent at 3:11–14 ("the nonaqueous electrolyte used in the nonaqueous secondary battery of the present invention contains a compound having at least two nitrile groups in the molecule."), '019 Patent at 3:21–23¹ (same); '446 Patent at 2:49-52 ("An object of the present invention is to provide a nonaqueous secondary battery having a high capacity, good charge discharge cycle characteristics and high storage characteristics."). The first passage describes that the "nonaqueous electrolyte used in the nonaqueous secondary battery ... contains a compound having at least two nitrile groups in the molecule," but does not further limit the nitrile weight. The second passage does not even mention the nitrile groups, let alone limit the nitrile weight. As such, the Court concludes that these passages, individually or collectively, do not limit the nitrile weight in the manner required by Defendant's proposed construction and that the "present invention" language does not function as a disclaimer.

Fourth, the Court agrees with Plaintiff that Defendant's proposed construction improperly limits the scope of the claim term to the disclosed embodiment. Liebel-Flarsheim, 358 F.3d at 913 ("[I]t is improper to read limitations from a preferred embodiment described in the specification—even if it is the only embodiment—into the claims absent a clear indication in the intrinsic record that the patentee intended the claims to be so limited.").

<sup>&</sup>lt;sup>1</sup> Defendant cites to '019 Patent at 3:24–35, but that passage does not use the phrase "present invention."

Therefore, for the reasons described above, the Court's final construction for these terms is plain-and-ordinary meaning.

E. Term #5: "0.97  $\le$ x<1.02, 0.8 $\le$ y<1.02, 0.002 $\le$ z $\le$ 0.05, and 0 $\le$ v $\le$ 0.05" / "0.97 $\le$ a<1.02, 0.8 $\le$ b<1.02, 0 $\le$ c $\le$ 0.02, and 0 $\le$ d $\le$ 0.02"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"0.97 \(\leq x < 1.02\), 0.8 \(\leq y < 1.02\), 0.002 \(\leq z \leq 0.05\), and 0 \(\leq v \leq 0.05\)" / "0.97 \(\leq a < 1.02\), 0.8 \(\leq b < 1.02\), 0 \(\leq c \leq 0.02\)"	No construction necessary; Plain and ordinary meaning	Indefinite
U.S. Patent No. 8,691,446 Patent, Cls. 1, 2, 3, 4; U.S. Patent No. 9,350,019, Cls. 1, 2; U.S. Patent No. 9,077,035, Cl. 1		

# **The Parties' Positions:**

Defendant contends there are two bases for indefiniteness: 1) Different numbers of significant figures at the range endpoints for specific variables and 2)  $M^2$  and  $M^3$  are described as being both optional and required. The formulas at issue are:

(1)

- $\bullet \quad Li_xM^1_yM^2_zM^3_vO_2$
- $\bullet \quad \text{Li}_{a}\text{M}^{1}{}_{b}\text{M}^{2}{}_{c}\text{M}^{3}{}_{d}\text{O}_{2} \tag{2}$

and the variable ranges are:

- $0.97 \le x < 1.02$
- 0.8≤y<1.02,
- 0.002≤z≤0.05
- 0≤v≤0.05

- $0.97 \le a < 1.02$
- 0.8≤b<1.02
- 0≤c≤0.02
- 0≤d≤0.02

Different numbers of significant figures at the range endpoints: Defendant contends that the ranges of y and b (0.8≤{y, b}<1.02) have 1 significant figure on the low end and 3 on the high end. Opening at 19. Defendant contends that a small change in the number at the low end (e.g., 0.75 versus 0.84, both of which round to 0.8) can make a huge difference, a POSITA would not understand what the range values are. *Id.* Defendant contends that the "absence of a clear range set forth with mathematical precision does not provide the POSITA with 'clear notice of what is claimed[.]'" *Id.* at 20 (quoting *Nautilus*, 572 U.S. at 899; citing *Astrazeneca AB v. Mylan Pharma*. *Inc.*, 2021 WL 5816742, at \*3 (Fed. Cir. Dec. 8, 2021) (emphasizing importance of mathematical precision, and construing percentages to five decimals)). Defendant contends that similar problems exist for z, c, v, and d. *Id.* at 19–20.

In its response, Plaintiff contends that a POSITA would understand the ranges. Response at 15. Plaintiff also contends that more significant figures are needed when the number is smaller. *Id.* Plaintiff contends that *AstraZeneca* does not support Defendant's argument because in that case, the Court reported that certain significant figures are amenable to construction. Response at 15 (citing *AstraZeneca*, 19 F.4th at 1335).

Plaintiff contends that "[t]o the extent [Defendant] and its expert contest the precision of the claimed ranges, this is not a question of indefiniteness, but one of construction." *Id.* at 16. Plaintiff contends that because "a POSITA would have no difficulty applying standard scientific convention to the claimed ranges[,]" under *Nautilus*, the ranges are defined with sufficiently reasonable certainty to apprise a POSITA of the scope of the claims. *Id*.

In its reply, Defendant first contends that Plaintiff's plain-and-ordinary construction is "wrong because it necessarily requires this Court to rewrite the claim to insert a word of approximation, such as 'about' or 'approximately,' where no such word exist." Reply at 7. Defendant contends that Plaintiff's expert's comments that "rounding" is "often" used actually supports indefiniteness as there are "conflicting interpretations" when rounding should be used. *Id.* (citing *UUSI*, *LLC v. United States*, 131 Fed. Cl. 244, 266 (2017)).

Defendant contends that the asymmetry in the precision the upper and lower ends of the range "injects ambiguity into the claim because a POSITA cannot determine the scope of the claims. For example, within the M<sup>2</sup> range in formula 1, a POSITA does not know whether the upper value for M<sup>2</sup> can be .054444 (out to a thousand places) followed by a 9." *Id.* at 8.

Defendant further contends that the "asymmetry of the formulas renders the claim indefinite because it creates a situation where  $M^2$ 's 'z' value in formula 1 can be less than its 'c' value in formula 2, contrary to the explicit teachings of the patents." *Id.* Defendant contends that this lack of precision can result in a case where c is ten times the value of z (0.02 v. 0.002), which is problematic as that the specification "requires" z > c. *Id.* (citing '446 Patent at 14:46–47). Defendant contends that if  $M^2$  is in greater amounts in formula (2) than in formula (1), "their function of storing and releasing Li ions is easily impaired." *Id.* at 8–9 (quoting '446 Patent at 14:37–41). According to Defendant, this indicates that Plaintiff's plain-and-ordinary construction is incorrect. *Id.* at 9.

With respect to Plaintiff's argument that "the level of precision necessary to define the relevant ranges is greater with respect to elements that are present in smaller quantities," Defendant

contends that that argument is nonsensical because the lower bounds of  $M^1$  and  $M^3$  only use one and zero numbers, respectively, after the decimal point (*i.e.*, 0.8 and 0, respectively) while  $M^2$  uses three numbers after the decimal point (*i.e.*, 0.002). *Id.* Defendant also contends that "nothing in the patents or [Plaintiff's expert's] testimony explains why there must be 3 decimal points level of precision on the low end of  $M^2$ , but not the top end of the  $M^2$  value." *Id.* 

In its sur-reply, Plaintiff contends that Defendant has not shown by clear and convincing evidence that the claims are indefinite. Sur-Reply at 8. Plaintiff contends that under Defendant's logic, "any claim having a limitation with a numerical value would be indefinite because the nature of numbers is that further precision is always possible through additional significant digits." *Id.* Plaintiff contends that logic is contrary to case law. *Id.* (quoting *BASF Corp. v. Johnson Matthey Inc.*, 875 F.3d 1360, 1365 (Fed. Cir. 2017) (holding that "reasonable certainty" "does not require absolute or mathematical precision") (internal quotation marks omitted)).

Plaintiff contends that Defendant's significant figures argument is a "red herring" as those numbers are simply the bounds of the claimed ranges, which was understood by Defendant's expert, and would be understood with "reasonable certainty" by a POSITA. *Id.* at 8–9.

With respect to Defendant's argument that a plain-and-ordinary meaning construction "reads out a preferred embodiment," Plaintiff contends that the possibility that a claim excludes a preferred embodiment does not render the claim to be indefinite. *Id.* at 9. Plaintiff further contends that there are "plenty of values for 'z' and 'c' that satisfy the claim limitations and are consistent with the preferred embodiment." *Id.* 

*M*<sup>2</sup> and *M*<sup>3</sup> are described as being both optional and required: Defendant contends that because the low end value for M<sup>2</sup> can be 0 in formula (2) and the low end value for M<sup>3</sup> can be 0 in both formula (1) and (2), they can be read out of the formulas altogether. Opening at 20. But Defendant also contends that the claims require both M<sup>2</sup> and M<sup>3</sup> being included. *Id.* Defendant further contends that nothing in the specification describes that M<sup>2</sup> and M<sup>3</sup> can be excluded. *Id.* at 21. Based on this, Defendant contends that because the claims simultaneously require the presence of M<sup>2</sup> and M<sup>3</sup> but also describe them being as optional, this is contradictory and thus indefinite. *Id.* at 20–21.

In its response, Plaintiff contends that the claims specifically allow the M<sup>2</sup> and M<sup>3</sup> to be optional, and that a POSITA would understand that. Response at 16. More specifically, Plaintiff contends that "because the subscripts in those particular instances include zero, a POSITA reading the claim would understand that M<sup>2</sup> in Formula (2) and M<sup>3</sup> in Formulas (1) and (2) are optional and do not need to be present[.]" *Id.* Plaintiff contends that "the claims allow for optionality, and thus are internally consistent and reconcilable to a POSITA." *Id.* at 17.

Plaintiff contends that the specification recites that 0 can be the lower bound for M<sup>3</sup>, and that the specification expressly says that a compound "may contain no M<sup>3</sup>." *Id.* at 17 (quoting '446 Patent at 14:2–6, '019 Patent at 14:9–13, '035 Patent at 9:20–24). Plaintiff also contends that the specification recites that 0 can be the lower bound for M<sup>2</sup>. *Id.* (citing '446 Patent at 11:19-20, '019 Patent at 11:27-28, '035 Patent at 6:33-34).

In its reply, Defendant concedes that the specification allows M<sup>3</sup> to be optional. Reply at 9. But Defendant contends that the specification does not recite that M<sup>2</sup> is optional, rather, it only says that there is "less necessity" for the compound to contain M<sup>2</sup>. *Id.* at 9–10.

Defendant also contends that the specification teaches away from M<sup>2</sup> being 0. *Id.* at 10. More specifically, Defendant contends that the specification describes that there needs to be a ratio of z to c, *e.g.*, 1.5:1. *Id.* But when c = 0, then the ratio is infinite which Defendant contends reads out an embodiment. *Id.* In particular, Defendant contends that the specification "consistently" requires M<sup>2</sup> where "two or more lithium-containing transition metal oxides having the different average particle sizes are used." *Id.* (citing '446 Patent at 11:40–42 ("when the amount of M<sup>2</sup> and the like added to the material (A) is relatively small, advantageous effects can be attained"); *see also* '446 Patent at 10:42–59, 14:17–21, and 14:54–15:6; '019 Patent at 10:37–38; 11:48-50; '035 pat. at 6:54-57; 10:16-17). Defendant contends that allowing M<sup>2</sup> to be optional excludes the embodiment in '446 Patent at 11:40–42. *Id.* 

Finally, Defendant contends that Plaintiff's expert provided contradictory statements during IPR and now. *Id.* at 10–11.

In its sur-reply, with respect to Defendant's argument that a plain-and-ordinary construction could read out a preferred embodiment, Plaintiff contends that this argument is inapposite because "the plain and ordinary meaning of the terms do not entirely read out the preferred embodiments." Sur-Reply at 9.

With respect to Defendant's argument that the specification does not describe that  $M^2$  is optional, but rather that there is "less necessity" for the compound to contain  $M^2$ , Plaintiff contends

that "less necessity" and the claim language are compatible and still support the conclusion that  $M^2$  is optional. Id.

Plaintiff also contends that because the parties agree  $M^3$  is not indefinite,  $M^2$  likewise is not as they have both have 0 as the lower bound. *Id.* at 9–10.

With respect to Defendant's contention that Plaintiff's expert provided contradictory statements during IPR and now, Plaintiff contends that Defendant mischaracterizes and takes statements out of context. *Id.* at 10.

### The Court's Analysis:

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff that because Defendant has not provided clear and convincing evidence that a POSITA would not understand the scope of this term with "reasonable certainty," the term is not indefinite and that the proper construction is plain-and-ordinary meaning for the reasons that follow.

Different numbers of significant figures at the range endpoints: First, the Court agrees that with Plaintiff that Defendant's argument that a term is indefinite simply because it does not have as many significant digits as another number is completely contrary to the Federal Circuit's guidance. More specifically, the Circuit has held that "reasonable certainty" "does not require absolute or mathematical precision" yet Defendant's position is that the term is indefinite because there is not enough mathematical precision in the endpoints of the range. BASF, 875 F.3d at 1365.

**Second**, the Court agrees with Plaintiff that Defendant's significant figures argument is a "red herring," as those numbers are simply the bounds of the claimed ranges. More precisely, the number of significant figures is a red herring because changing the precision does not change the claim scope, e.g., changing  $0.002 \le z \le 0.05$  to  $0.002 \le z \le 0.050$ , despite increasing the number of digits after the decimal place. Rather, a POSITA understand that 0.05 and 0.050 are mathematically equal, regardless of the the number of digits after the decimal place.

**Third**, the Court agrees with Plaintiff that under Defendant's logic, any claim having a limitation with a numerical value would be indefinite because further precision is always possible through additional significant digits.

*Fourth*, the Court agrees with Plaintiff that even if a claim excludes a preferred embodiment, that does not render the claim to be indefinite.

 $M^2$  and  $M^3$  are described as being both optional and required: First, the parties appear to agree that the specification describes  $M^3$  as optional. Response at 17, Reply at 9. **Second**, the Court agrees with Plaintiff that if  $M^3$  is not indefinite, then  $M^2$  should not be either.

*Third*, both parties agree there is no ambiguity in the claim language itself and both parties agree that the claims allow for M<sup>2</sup> not to be present in the claimed compound. Opening at 16, Response at 16. Therefore, the Court concludes that a POSITA would understand with "reasonable certainty" the meaning of this claim term.<sup>2</sup>

<sup>&</sup>lt;sup>2</sup> While the specification does not expressly recite that M<sup>2</sup> is optional, the Court does not consider that to be § 112, ¶ 2 indefiniteness issue, but at most a § 112, ¶ 1 written description issue. The Court does not take a position whether 35

**Fourth**, the Court agrees with Plaintiff that even if a claim excludes a preferred embodiment, that does not render the claim to be indefinite.

Construction: Because the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning and because Defendant does not allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning, the Court concludes that the term should be construed as having its plain-and-ordinary meaning. Azure Networks, 771 F.3d at 1347; Thorner, 669 F.3d at 1365.

Therefore, for the reasons described above, the Court finds that the term is not indefinite and should be construed according to its plain-and-ordinary meaning.

F. Term #6: "M<sup>2</sup> represents Mg, or Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn, ... [and] a content of Mg in the formula (1) is from 0.15% by mole to less than 2% by mole based on an amount of the metal M<sup>1</sup>"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
	Construction	Constituction
	No construction necessary;	Indefinite
at least one metal element	Plain and ordinary meaning	
selected from the group		
consisting of Ti, Zr, Ge, Nb,		
Al and Sn, [and] a content		
of Mg in the formula (1) is		
from 0.15% by mole to less		

there is a written description issue. Rather, the Court only raises the written description requirement to categorize Defendant's argument as appearing to be directed towards written description than indefiniteness.

than 2% by mole based on an amount of the metal M <sup>1</sup> "	
U.S. Patent No. 9,350,019, Cl. 1	

### The Parties' Positions:

Defendant contends that this term is indefinite because there is a broad range  $(0.002 \le z \le 0.05)$  and a narrow range ("from 0.15% by mole to less than 2%") in the claim and that is indefinite per MPEP § 2173.05(c) ("Numerical Ranges and Amounts Limitations"). Opening at 22. More specifically, Defendant contends that "M² can be magnesium alone or in combination with 'at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn." *Id.* (quoting '019 Patent, Claim 1, Limitation [c]). Defendant contends that, later in the claim, "the amount of magnesium is capped by a ratio of '0.15% by mole to less than 2% by mole based on an amount of the metal M¹." *Id.* (quoting '019 Patent, Claim 1, Limitation [d]). Defendant contends that this claim language renders the claim indefinite from a POSITA's perspective. *Id.* 

Defendant contends that Examiner flagged this issue during prosecution and Applicant canceled the claims, but sought the same exact claim limitation later which was ultimately granted. *Id.* at 22–23.

In its response, Plaintiff contends that the term is not indefinite "because its scope is clear, especially in light of the intrinsic evidence" and that its expert agrees. Response at 18.

Plaintiff contends that these two ranges, while related, "are wholly separate and can exist without conflict." Response at 19 (citing ECF No. 52-1 (Lucht expert declaration) at ¶ 64). In particular, Plaintiff contends that:

The requirement that the subscript for  $M^2$  is  $0.002 \le z \le 0.05$  describes the amount of  $M^2$  with respect to **the entire lithium-containing transition metal oxide formula**. The requirement that 'a content of Mg in the formula (1) is from 0.15% by mole to less than 2% by mole based on an amount of the metal  $M^1$ ' describes the amount of Mg in the entire lithium-containing transition metal oxide with respect to the amount of metal in  $M^1$ .

*Id.* (emphasis in Plaintiff's brief, internal citations removed). In other words, Plaintiff contends that "the two claimed ranges describe different limitations of the claim, based on different features. Accordingly, the [] limitation, as written, does not claim a range within a range." *Id.* (internal citations omitted).

Plaintiff contends that Defendant's MPEP argument also fails because it pertains to when the boundaries of the claim are not discernable. *Id.* In particular, Plaintiff points out the examples given in the MPEP, *e.g.*, "(A) a temperature of between 45 and 78 degrees Celsius, preferably between 50 and 60 degrees Celsius" does not make clear what the claimed range is. *Id.* at 19–20. Plaintiff contends that that example is indefinite because it is unclear which range (45 to 78°C or 50 to 60°C) is being claimed, but here "the limitation at issue clearly indicates that Mg must be present, and in what quantities."

In its reply, Defendant contends that Plaintiff's argument is a "strained" reading of the claim and not how a POSITA would understand it. Reply at 11. Defendant then otherwise repeats its indefiniteness and prosecution arguments that it provided in its opening brief. *Id.* at 12.

In its sur-reply, Plaintiff contends that Defendant repeats its prior arguments and that the "great flaw" in Defendant's argument is that it relies on the prosecution history for the '446 Patent when this term does not appear in the '446 Patent, but only in the '019 Patent. Sur-Reply at 11. Plaintiff contends that this term does not require a narrower range be within a broader range and that is why Examiner allowed the claims. *Id*.

### The Court's Analysis:

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff that because Defendant has not provided clear and convincing evidence that a POSITA would not understand the scope of this term with "reasonable certainty," the term is not indefinite and that the proper construction is plain-and-ordinary meaning.

The Court agrees with Plaintiff that this term does not require a narrower range within a broader range. More specifically, Claim 1 of the '019 Patent requires that  $M^2$  represents Mg, or Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn. Claim 1 also limits  $M^2$  by describing that its molar ratio is between  $0.002 \le z \le 0.05$  and that the content of Mg is 0.15% by mole to 2% by mole based on an amount of metal element  $M^1$ . The two numerical relationships are independent of each other, insofar as they are based on different things. More specifically, the subscript for  $M^2$  describes the amount of  $M^2$  with respect to the entire lithium-containing transition metal oxide formula. By contrast, the "content of Mg in the formula (1) is from 0.15% by mole to less than 2% by mole based on an amount of the metal  $M^1$ " is with respect to the amount of metal in  $M^1$ . Because these two numerical relationships are based

on different denominators, the Court concludes there is no conflict and thus the term is not indefinite.

Because the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning and because Defendant does not allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning, the Court concludes that the term should be construed as having its plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347; *Thorner*, 669 F.3d at 1365.

Therefore, for the reasons described above, the Court finds that the term is not indefinite and should be construed according to its plain-and-ordinary meaning.

### G. Term #7: "at least 3.5 g/cm3"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"at least 3.5 g/cm <sup>3</sup> "  U.S. Patent No. 8,691,446  Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cls. 1; U.S.  Patent No. 9,077,035, Cl. 7		Indefinite

### **The Parties' Positions:**

Defendant contends that this is indefinite because there is no upper-bound specified in the claims for this term. Opening at 24. Defendant contends that the specification describes that the preferred upper limit is 4.6 g/cm<sup>3</sup>. *Id.* (citing '446 Patent at 7:57–67). Defendant further contends that the specification describes that battery reliability decreases as density increases. *Id.* Given

that, Defendant contends that without specifying an upper-bound in the claims, the claims lack enablement. *Id.* 

In its response, Plaintiff contends that enablement is a question of validity, which is not appropriate to resolve during claim construction. Response at 20. Plaintiff also contends that the term is understandable on its own and that Defendant's expert understands it. *Id.* Plaintiff contends that Defendant confuses breadth with indefiniteness. *Id.* at 21. Plaintiff also contend that the Federal Circuit has held that "[o]pen-ended claims are not inherently improper; as for all claims their appropriateness depends on the particular facts of the invention, the disclosure, and the prior art." *See Andersen Corp. v. Fiber Composites, LLC*, 474 F.3d 1361, 1376-77 (Fed. Cir. 2007).

In its reply, Defendant again contends that because the claim does not specify an upper limit, a POSITA would not understand the scope of the claim term with "reasonable certainty" given that a POSITA would understand—and the specification discloses—that battery reliability decreases as density increases. Reply at 12–13.

In its sur-reply, Plaintiff again contends that failure to provide an upper limit has "no support in law or fact." Sur-Reply at 11. Plaintiff contends that "both experts agree that the term 'at least 3.5 g/cm<sup>3</sup>' is clear." *Id.* at 12.

### The Court's Analysis:

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff that because Defendant has not provided clear and convincing evidence that a

POSITA would not understand the scope of this term with "reasonable certainty," the term is not indefinite and that the proper construction is plain-and-ordinary meaning for the reasons that follow.

First, given there is no dispute that a POSITA would understand that the claim term "at least 3.5 g/cm³" simply means "greater than or equal to 3.5 g/cm³. Second, the Court concludes that a lack of an upper bound does not render the term to be indefinite and Defendant has not cited any authority for that proposition. At best, Defendant attempts to conflate enablement with indefiniteness, in order to bolster its indefiniteness case. Third, the Court agrees with Plaintiff that Defendant confuses breadth with indefiniteness. Based on these reasons, the Court concludes that Defendant has failed to provide clear-and-convincing that a POSITA would understand with "reasonable certainty" the scope of this claim term.

Because the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning and because Defendant does not allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning, the Court concludes that the term should be construed as having its plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347; *Thorner*, 669 F.3d at 1365.

Therefore, for the reasons described above, the Court finds that the term is not indefinite and should be construed according to its plain-and-ordinary meaning.

<sup>&</sup>lt;sup>3</sup> As was the case for note 2, the Court does not take a position whether there is an enablement issue. Rather, the Court only raises the enablement requirement to categorize Defendant's argument as appearing to be directed towards enablement than indefiniteness.

H. Term #8: "M¹ represents at least one transition metal element selected from Co, Ni and Mn, . . . wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
"M¹ represents at least one transition metal element selected from Co, Ni and Mn, wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole"	No construction necessary; Plain and ordinary meaning	Indefinite
U.S. Patent No. 9,077,035, Cl. 1		

### The Parties' Positions:

Defendant contends that this term is indefinite because it is unclear whether Co has to be selected or is merely optional. Opening at 24–25. More specifically, Defendant contends that the first part of the claim requires that at least one of Co, Ni and Mn be selected as M<sup>1</sup>. Opening at 25. As such, M<sup>1</sup> may be Co (either alone or together with Ni and/or Mn) or it may not include Co. *Id.* Defendant contends that the "wherein the content of Co in the transition metal M<sup>1</sup> of the formulae (1) and (2) is from 30% by mole to 100% by mole" requires that Co be M<sup>1</sup> and be 30% to 100% of the weight. *Id.* Based on the understanding of these two requirements, Defendant contends that the first part of the claim describes that Co is optional while the second half of the claim recites that Co is required. *Id.* Based on these conflicting requirements with respect to the optionality of Co, Defendant contends that the claim is indefinite. *Id.* 

In its response, Plaintiff contends that the first part of the claim "constitutes a *Markush* group wherein M<sup>1</sup> represents Co, or Co and at least one transition metal element selected from the group consisting of Ni and Mn." Response at 22. Plaintiff contends that the second part of the claim "simply confirms that the presence of Co is required." *Id*.

Plaintiff contends that the prosecution history supports its position as Co was in the originally filed claims. *Id.* Plaintiff then contends that Applicant amended the claim to create *Markush* group by adding Ni and Mn as additional elements and to require that Co is 30% to 100%. Response at 22–23 (quoting Response, Ex. 18 at 2–3). Plaintiff further contends that Applicant distinguished the pending claim from prior art by arguing that "Oda et al. JP '507 does not describe or teach such a content of Co, since Oda et al. JP '507 predominantly uses Ni as a transition metal." *Id.* at 23 (quoting Response, Ex. 18 at 10).

Plaintiff contends that "a POSITA would understand, that the content of Co in the transition metal M<sup>1</sup> of the formulae (1) and (2) is from 30% by mole to 100% by mole, and that M<sup>1</sup> may also include Ni and/or Mn." *Id*.

In its reply, Defendant contends that Plaintiff's assertion that the claim "constitutes a *Markush* group wherein M¹ represents [1] Co, or [2] Co and at least one transition metal element selected from the group consisting of Ni and Mn" is incorrect. Reply at 13 (quoting Response at 22 (annotations added)). More specifically, Defendant contends that the claim language does not recite two options as annotated, but rather only describes a single option, "M¹ represents at least one transition metal element selected from Co, Ni and Mn." *Id.* Defendant contends that had Applicant wanted to draft a claim in the manner Plaintiff argued, Applicant knew how to do so as

Claim 1 of the '019 Patent was drafted to allow for two options. '019 Patent, Claim 1, Limitation [c] ("...M² represents Mg, or Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn..."). Defendant contends that the contradictory nature of the claim language renders this claim term to be indefinite. *Id*.

In its sur-reply, Plaintiff contends that Defendant's indefiniteness argument is based on a "willful refusal to read the two parts of the term together and a refusal to consider how a POSITA would understand the effect of the latter portion of the term defining the Co content on the former portion of the term broadly defining the possible elements." Sur-Reply at 12. Plaintiff contends that "standard practice in drafting patent claims for some aspect to be defined more broadly and then further narrowed through specific limitations," in this case, Applicant first broadly drafted a *Markush* group to include Co and then narrowed it to require a specific amount of Co. *Id.* Plaintiff contends that a POSITA would understand these two limitations with reasonable certainty and that they are not irreconcilable. *Id.* at 12–13.

### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Defendant that the term is indefinite for the reasons that follow. *First*, the Court concludes that the plain language of the claim recites a contradiction. The first part of the claim recites a *Markush* group where Co is not necessarily required to be in the claimed compound while the second part of the claim recites that Co is necessarily required. For an element to simultaneously be optional and required is a contradiction on its face. As such, the Court concludes that there is

clear-and-convincing evidence that a POSITA would not understand with "reasonable certainty" what this claim requires.

Second, the Court disagrees with Plaintiff's interpretation of the claim. Plaintiff argues that the claim "constitutes a *Markush* group wherein M¹ represents [1] Co, or [2] Co and at least one transition metal element selected from the group consisting of Ni and Mn." Reply at 13 (annotations added). But rather than describe two options where Co is part of both options, the claim ("M¹ represents at least one transition metal element selected from Co, Ni and Mn") recites a *Markush* group where there are up to seven options (Co, Ni, Mn, Ni & Mn, Co & Mn, Co & Ni, Co & Ni & Mn) where Co is only present in four of the seven options. Therefore, Plaintiff's assertion that Co is necessarily present in the claimed compound is incorrect, which means that aforementioned contradiction exists.

Third, the Court disagrees with Plaintiff that Claim 1 simply follows standard drafting practice by reciting a broad limitation and then reciting another limitation that narrows the broad limitation. As described above, the first part of the claim recites a broad claim with seven options, but then the second part of the claim recites a narrowing limitation that does not apply to all options. In other words, the second part of the claim does not narrow the first part of the claim as it does not apply to some Markush group combinations. Therefore, the Court concludes that Plaintiff's interpretation that Claim 1 recites a broad limitation that is narrowed by another limitation is incorrect.

Fourth, the Court agrees with Defendant that had Applicant wanted to draft a claim in the manner Plaintiff argued, Applicant knew how to do so. More specifically, Claim 1 of the '019

Patent allows for two options (both of which required one particular element, Mg). '019 Patent, Claim 1, Limitation [c] ("...M² represents Mg, or Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn..."). But, despite knowing how to draft a claim that allow for two options, both of which could have included Co, Plaintiff chose not to do so for this claim term, thus supporting the Court's conclusion of indefiniteness.

Therefore, for the reasons described above, the Court finds that the term is indefinite.

I. Term #9: "the shutdown layer has a thickness A ( $\mu$ m) of 5 to 30, the heat-resistant layer has a thickness B ( $\mu$ m) of 1 to 10, a sum of A and B is 6 to 23, and a ratio A/B is 1/2 to 4"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
#9: "the shutdown layer has a thickness A (μm) of 5 to 30, the heat-resistant layer has a thickness B (μm) of 1 to 10, a sum of A and B is 6 to 23, and a ratio A/B is 1/2 to 4"  U.S. Patent No. 9,166,251, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite

### **The Parties' Positions:**

The formulas that are relevant to the indefiniteness dispute are:

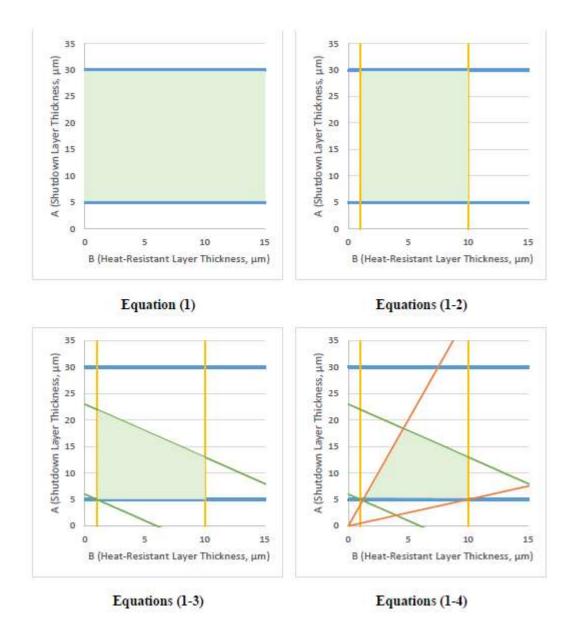
- $\bullet \quad 5 \le A \le 30 \tag{1}$
- $\bullet \quad 1 \le B \le 10 \tag{2}$
- $6 \le (A + B) \le 23$  (3)
- $\bullet \quad 1/2 \le A/B \le 4 \tag{4}$

where A and B are thicknesses of the shutdown layer and heat-resistant layer, respectively.

Defendant contends that this claim term is indefinite because A and B cannot, *e.g.*, both be at their maximum (*i.e.*, A=30 and B=10), without violating the  $6 \le A+B \le 23$  requirement. Opening at 26. Defendant contends that this is "internally inconsistent and logically impossible." *Id.* Defendant contends that the  $1/2 \le A/B \le 4$  limitation "amplifies" the problem, but does not appear to explain why. *Id.* at 27. Defendant contends that "[a]s a question of algebra, these four equations are cannot be reconciled," and thus the claim term is indefinite. *Id.* (citing *Fargo Elecs. Inc. v. Iris, Ltd.*, 287 F. App'x 96, 100 (Fed. Cir. 2008)).

Defendant contends that Applicant added ranges for A and B in order to added to get around prior art, which disclosed ranges of 5–35 µm for A and 1–16 µm for B. *Id.* at 28.

Plaintiff contends that there is no inconsistency and that it is a simple matter for determine what values of A and B satisfy the equations and which do not. Response at 24–25. Plaintiff contends that the claim does not require that each equation be met in isolation, but rather values of A and B need to meet all four equations simultaneously. *Id.* at 25. Plaintiff argues that each limitation successively narrows the scope of the claim to what infringes and what does not. Response at 25–26. Plaintiff provides the following illustration to show how each equation further limits the claim scope of what are acceptable values for the shutdown layer and heat-resistant layer thicknesses. Response at 26.



Based on this, Plaintiff contends that rather than being indefinite, "a POSITA would readily understand that the four Equations define a discrete set of values for A and B, and thus describe with mathematical precision the allowed values, far exceeding the reasonable certainty required by *Nautilus*." *Id.* at 26–27.

In its reply, Defendant contends that Plaintiff is asking the Court to rewrite the claim in order to preserve validity. Reply at 14. Defendant also contends that Plaintiff has not explained why the claim limitation provides that the shutdown layer (A) can have a thickness up to 30  $\mu$ m. *Id*.

In its sur-reply, Plaintiff contends that a POSITA would be able to determine which values of A and B infringe and which do not. Sur-Reply at 13. Plaintiff contends that Defendant's expert admits that given values for A and B, he would be able to determine whether that pair of values fell within the claim scope. *Id.* (citing Sur-Reply, Ex. 19 at 116:13–118:14). Plaintiff also contends that the figures it provided above "are instructive charts that describe a mathematically bounded set of values for A and B that meet all four limitations." *Id.* 

With respect to why shutdown layer (A) can have a thickness up to 30 µm, Plaintiff contends that is the wrong question to ask and all the matter is whether a POSITA could determine the set of values of A and B that fall within the claim scope with reasonable certainty. *Id.* at 14.

### The Court's Analysis:

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff that because Defendant has not provided clear and convincing evidence that a POSITA would not understand the scope of this term with "reasonable certainty," the term is not indefinite and that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, the Court concludes that there is no ambiguity in the above formulas that would prevent a POSITA from determining whether a specific pair of values for the thicknesses of the

shutdown layer and heat-resistant layer (*i.e.*, A and B, respectively) meet all four equations. Plaintiff's charts above depict the exact set of values for A and B that meet all four equations. Furthermore, Defendant's expert concedes that he would able to determine whether a set of values for A and B met all four equations.

**Second**, Defendant's core argument appears to be that when a specific value of A and/or B meets one equation, but is mathematically impossible to also meet other equations, the claim is indefinite. Defendant contends that  $A = 30 \mu m$  illustrates its point as while that value of A meets equation (1) ( $5 \le A \le 30$ ), it cannot possibly meet equation (3) ( $6 \le (A + B) \le 23$ ). But the Court is not persuaded by Defendant's argument because Defendant conflates what are non-infringing values with indefiniteness.

All that is required for a claim to be not indefinite is for a POSITA to understand with "reasonable certainty" what scope of the invention is. *Nautilus*, 572 U.S. at 910. As described above, a POSITA would understand the scope of this term with reasonable certainty. But the fact that some values of A and B meet some equations (*i.e.*, limitations), but not others, is a classic case of non-infringement. If anything, because the claim is written in terms of mathematical relationships, the scope of the claim is crystal clear—which means it is not indefinite—such that it is very easy to determine what values of A and B fall outside of the scope of the claim—which means that they do not infringe.

*Third*, with respect to Defendant's argument that Plaintiff is asking the Court to redraft the claims, the Court rejects this argument because Defendant has not explained how plain-and-ordinary meaning somehow redrafts the claim. Furthermore, as described above, the scope of the

claim is so clear that even Defendant's expert can determine what values of A and B will infringe and which do not and that Plaintiff can provide a colorful graphic to pictorially depict the claim scope. Therefore, the Court does not need to redraft the claim to preserve its validity (nor does it have the power to do so).

Because the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning and because Defendant does not allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning, the Court concludes that the term should be construed as having its plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347; *Thorner*, 669 F.3d at 1365.

Therefore, for the reasons described above, the Court finds that the term is not indefinite and should be construed according to its plain-and-ordinary meaning.

### J. Term #10: "a particle size of 0.2 $\mu m$ or less . . . a particle size of 2 $\mu m$ or more"

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction
#10: "a particle size of 0.2 μm or less a particle size of 2 μm or more"  U.S. Patent No. 9,166,251, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite

### **The Parties' Positions:**

Defendant first contends that this term is indefinite because the patentee did not specify the x, y, z dimensions of the particle size. Opening at 28. Defendant cites to extrinsic evidence to 52

illustrate that the particle size could be the diameter at the largest/smallest point, average length/width/height, etc. *Id.* at 29. Defendant contends that because "particle size' could mean diameter at largest point, diameter at smallest point, or average length/width/height, the claim does not set forth the metes and bounds of the invention and hence is indefinite." *Id.* 

In its response, Plaintiff contends that Defendant's expert does not address Defendant's claim that a POSITA would be unable to determine what "particle size" means in the context of the '251 Patent. Response at 28. Plaintiff contends that this omission alone is fatal to Defendant's indefiniteness argument. *Id*.

Plaintiff contends that the specification addresses the irregularity of particles and how to measure them (as the diameter of a hypothetical sphere using a laser diffraction particle analyzer). *Id.* at 28–29. Plaintiff contends that "[1]aser diffraction particle sizing is a well-known technique that results in particle size distributions reported as volume equivalent sphere diameters." *Id.* at 29. Plaintiff also points to one of Defendant's exhibits that describes using laser diffraction to measure volume equivalent sphere diameter. *Id.* at 29 (quoting Opening, Ex. 16 at 14–15).

In its reply, Defendant contends that specification does not describe how to measure the "proportion" of the small/large particles. Reply at 15. Defendant contends that while the specification describes using the "Horiba" device to measure the "average particle size of all particles in a layer by creating a volumetric equivalence of all particles and dividing by the total number of particles, there is nothing in the patent that says this device should be used to measure the size of a 'proportion' of particles." *Id*.

In its sur-reply, Plaintiff first contends that the Defendant's new argument is waived because it was not raised until Reply. Sur-Reply at 14 (quoting *United States v. Jackson*, 426 F.3d 301, 304 n.2 (5th Cir. 2005) ("Arguments raised for the first time in a reply brief... are waived.")).

Plaintiff next contends that Defendant "is wrong on the operation of the Horiba device; it does not provide a single average particle size across the entire layer but rather provides a distribution of particle sizes." Sur-Reply at 15 (citing Sur-Reply, Ex. 20 at 68–69; Opening, Ex. 15 at 4).

Plaintiff finally contends that "[t]he specific capabilities of the Horiba device aside, [Defendant's] argument has nothing to do with whether the claim term itself is indefinite." Sur-Reply at 15. Plaintiff contends that the issue is that "[t]he claim requires that the proportion of the particles that are 0.2 µm or less, for example, make up 10 vol % or less of the layer" and "[t]here is no dispute of how to interpret the meaning of having a proportion of particles in the heat-resistant layer having a particular particle size." *Id*.

### **The Court's Analysis:**

After reviewing the parties' arguments and considering the applicable law, the Court agrees with Plaintiff that because Defendant has not provided clear and convincing evidence that a POSITA would not understand the scope of this term with "reasonable certainty," the term is not indefinite and that the proper construction is plain-and-ordinary meaning for the reasons that follow. *First*, none of Defendant's arguments have anything to do with indefiniteness. Rather,

Defendant's arguments appear to be directed towards enablement.<sup>4</sup> *Second*, Defendant does not appear to provide an expert declaration that explains why a POSITA would not understand, with "reasonable certainty," what the scope of the claimed invention is. Based on these two reasons, the Court finds that Defendant has not provided clear-and-convincing evidence that the claim term is indefinite.

Third, Defendant's basis for its indefiniteness allegation shifted from its Opening brief to its Reply brief, which indicates, at minimum, that those arguments are not strong. More specifically, Defendant abandoned its Opening indefiniteness argument in its Reply brief which indicates that even Defendant realized there was not much merit to that argument. Defendant first raised its Reply indefiniteness argument in its Reply brief, which either indicates that Defendant did not think that it was as strong as its Opening indefiniteness argument or that Defendant only thought of it for the Reply brief, neither of which indicates that Defendant's Reply indefiniteness argument is strong.

Because the "heavy presumption" is that terms should be construed according to their plain-and-ordinary meaning and because Defendant does not allege lexicography or disclaimer, which are the only two exceptions to the general rule that a term should be construed as having its plain-and-ordinary meaning, the Court concludes that the term should be construed as having its plain-and-ordinary meaning. *Azure Networks*, 771 F.3d at 1347; *Thorner*, 669 F.3d at 1365.

<sup>&</sup>lt;sup>4</sup> As was the case for note 2, the Court does not take a position whether there is an enablement issue. Rather, the Court only raises the enablement requirement to categorize Defendant's argument as appearing to be directed towards enablement than indefiniteness.

Therefore, for the reasons described above, the Court finds that the term is not indefinite and should be construed according to its plain-and-ordinary meaning.

### IV. CONCLUSION

In conclusion, for the reasons described herein, the Court adopts the below constructions as its final constructions.

**SIGNED** this 10th day of November 2022.

ALAN D ALBRIGHT

UNITED STATES DISTRICT JUDGE

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction	Court's Final Construction
#1: "positive electrode mixture layer" U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,077,035, Cl. 1	No construction necessary; Plain and ordinary meaning	"A mixture of at least two lithium-containing transition metal oxides formed on one or both sides of an electrode current collector"	Plain-and-ordinary meaning
#2: "the positive electrode contains, as [an] active material[s], at least two lithium-containing transition metal oxides having different average particle sizes."  U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cl. 1; U.S. Patent No. 9,350,019, Cl. 1; U.S.	No construction necessary; Plain and ordinary meaning	"The active material of the positive electrode is a mixture formed from two or more lithium-containing transition metal oxides, at least two of the transition metal oxides having different average particle sizes"	Plain-and-ordinary meaning

Term	Plaintiff's Proposed Construction	Defendant's Proposed Construction	Court's Final Construction
#3: "different compositions of elements" / "is different from"	No construction necessary; Plain and ordinary meaning	"The first and second lithium- containing transition metal oxides do not share all of the	Plain-and-ordinary meaning
U.S. Patent No. 8,691,446 Patent, Cls. 1, 2, 3 U.S. Patent No. 9,350,019, Cls. 3, 4		same chemical elements"	
#4: "a compound having at least two nitrile groups"	No construction necessary; Plain and ordinary meaning	"A compound with at least two nitrile groups that are 1% or less by total weight of the	Plain-and-ordinary meaning
U.S. Patent No. 8,691,446 Patent, Cls. 1, 3; U.S. Patent No. 9,350,019, Cls. 1		electrolyte"	
#5: "0.97 < x<1.02, 0.8 0.8 and 0	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and- ordinary meaning.
U.S. Patent No. 8,691,446 Patent, Cls. 1, 2, 3, 4; U.S. Patent No. 9,350,019, Cls. 1, 2; U.S. Patent No. 9,077,035, Cl. 1			
	i		

Term	Plaintiff's Proposed	Defendant's Proposed	Court's Final	ıal
	Construction	Construction	Construction	on
#6: "M² represents Mg, or Mg   No	construction necessary;	Indefinite	Not indefinite.	Plain-and-
and at least one metal element Plain and ordinary meaning	Plain and ordinary meaning		ordinary meaning.	
selected from the group consisting of Ti. Zr. Ge. Nb. Al				
and Sn, [and] a content of				
Mg in the formula (1) is from				
U.15% by mole to less than 2%				
by mole based on an amount of the metal $M^{1,r}$				
U.S. Patent No. 9,350,019, Cl.				
1				
#7: "at least 3.5 g/cm <sup>3</sup> "	ury;	Indefinite	Not indefinite.	Plain-and-
	Plain and ordinary meaning		ordinary meaning.	
U.S. Patent No. 8,691,446				
Patent, Cls. 1, 3; U.S. Patent				
No. 9,350,019, Cls. 1; U.S.				
Patent No. 9,077,035, Cl. 7				

Term	Plaintiff's Proposed	Defendant's Proposed	Court's Final
	Construction	Construction	Construction
#8: "M¹ represents at least one transition metal element selected from Co, Ni and Mn, wherein the content of Co in the transition metal M¹ of the formulae (1) and (2) is from 30% by mole to 100% by mole."  U.S. Patent No. 9,077,035, Cl. 1	No construction necessary; Plain and ordinary meaning	Indefinite	Indefinite
#9: "the shutdown layer has a thickness A (μm) of 5 to 30, the heat-resistant layer has a thickness B (μm) of 1 to 10, a sum of A and B is 6 to 23, and a ratio A/B is ½ to 4"  U.S. Patent No. 9,166,251, CI.	No construction necessary; Plain and ordinary meaning	Indefinite	Not indefinite. Plain-and- ordinary meaning.

Term	Plaintiff's Proposed	Defendant's Proposed	Court's Final
	Construction	Construction	Construction
	No construction necessary;	Indefinite	Not indefinite. Plain-and-
or less a particle size of 2	Plain and ordinary meaning		ordinary meaning.
μm or more"			
U.S. Patent No. 9,166,251, Cl.			
1			



US009077035B2

## (12) United States Patent Kita et al.

(10) Patent No.:

US 9,077,035 B2

(45) **Date of Patent:** 

Jul. 7, 2015

### (54) NONAQUEOUS SECONDARY BATTERY AND METHOD OF USING THE SAME

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Tsubata, Ibaraki (JP); Hiroyasu Inoue,

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(JP)

(\*) Notice: Subject to any disclaimer, the term of this

patent is extended or adjusted under 35

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H01M 10/0567; Y02E 60/122 USPC ....... 429/231.1, 231.3, 231.6, 231.5, 340,

429/341, 50

See application file for complete search history.

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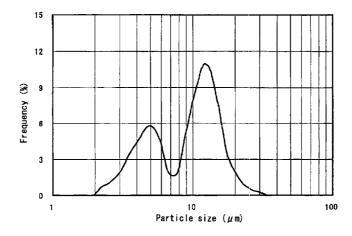
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### (57) ABSTRACT

A nonaqueous secondary battery containing a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electrolyte, wherein the positive electrode comprises, as active materials, two or more lithium-containing transition metal oxides having different average particle sizes, and the lithium-containing transition metal oxide having the smallest average particle size contains one or more of Mg, Ti, Zr, Ge, Nb, Al and Sn.

### 11 Claims, 3 Drawing Sheets



# US 9,077,035 B2 Page 2

(51) Int. Cl.  H01M 6/16 (2006.01)  H01M 10/0567 (2010.01)  H01M 4/131 (2010.01)  H01M 10/0569 (2010.01)  H01M 10/0525 (2010.01)  H01M 4/02 (2006.01)  (52) U.S. Cl.	JP 2002-42890 A 2/2002 JP 2004-63269 A 2/2004 JP 2004-182564 A 7/2004 JP 2004-259675 A 9/2004 JP 2004-349132 A 12/2004 JP 2005-5208 A 1/2005 JP 2005-93238 A 4/2005 JP 2005-285720 A 10/2005 JP 2005-302507 A 10/2005  OTHER PUBLICATIONS			
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Appx0120

Fig. 1

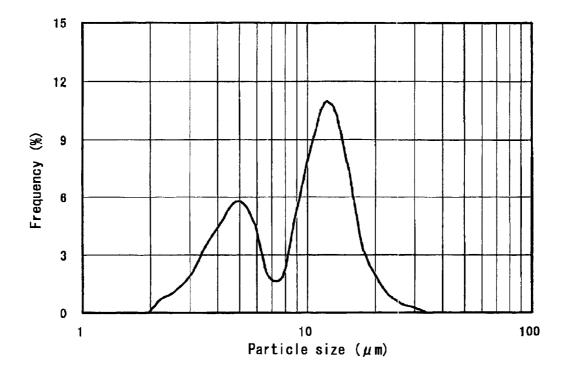


Fig. 2A

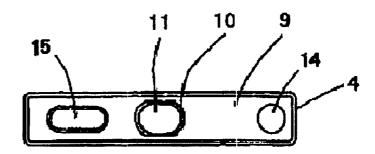


Fig. 2B

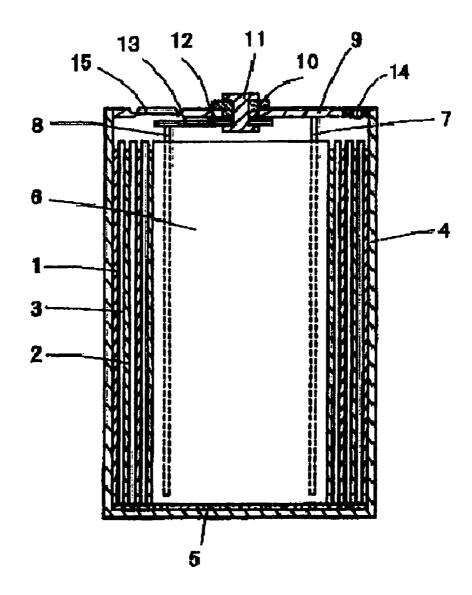
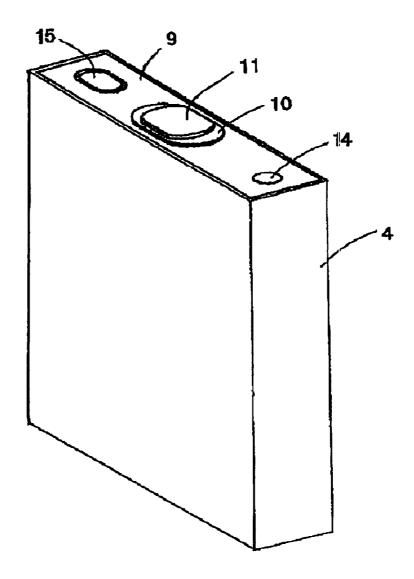


Fig. 3



# NONAQUEOUS SECONDARY BATTERY AND METHOD OF USING THE SAME

#### FILED OF THE INVENTION

The present invention relates to a nonaqueous secondary battery and a method of using the same.

## RELATED ART

In recent years, the secondary battery is an indispensable, important device as a power source of a personal computer or a cellular phone, or a power source for an electric vehicle or an electric power storage.

In particular, in applications for a mobile communication device such as a portable computer and a personal digital assistant, the battery is required to be made smaller and to trim weight. Under the current circumstances, however, the system of the battery is not easily made compact or lightweight, since an electric power consumed by a back light of a liquid crystal display panel or consumed to control the drawing of graphics is large, or the capacity of a secondary battery is not sufficiently large. In particular, a personal computer is progressively multi-functionalized by mounting a digital versatile disc (DVD) drive and so on. Thus, the power consumption thereof tends to increase. For this reason, it is highly required to increase the electric capacity of a secondary battery, in particular, the discharge capacity, when the voltage of a single battery is 3.3 V or higher.

Attention is paid to electric vehicles, which discharge no exhaust gas and make less noise in association with the increase of global environmental problems. Recently, hybrid electric vehicles (HEV), which adopt a system of storing regenerative energy generated at the time of braking in a battery and making effective use of the energy, or using an electric energy stored in a battery at the time of engine starting to increase the efficiency of the engine system, have gained popularity. However, since the electric capacity of the currently used battery is small, a plurality of batteries should be used to generate a sufficient voltage. For this reason, problems such that a space in the vehicle should be made smaller and that the stability of the vehicle body deteriorates arise.

Among secondary batteries, a lithium secondary battery 45 using a nonaqueous electrolyte attracts attention, since it generates a high voltage, has a lightweight and is expected to achieve a high energy density. In particular, a lithium secondary battery disclosed in JP-A-55-136131, in which a lithium-containing transition metal oxide, for example, LiCoO<sub>2</sub>, is 50 used as a positive electrode active material, and metal lithium is used as a negative electrode active material, is expected to attain a high energy density, since it has an electromotive force of 4 V or higher.

However, at present, in the case of a LiCoO<sub>2</sub> based secondary battery which uses LiCoO<sub>2</sub> as a positive electrode active material and a carbonaceous material such as graphite as a negative electrode active material, a charge final voltage thereof is usually 4.2 V or less. According to this charging condition, the charge capacity is only about 60% of the theoretical capacity of LiCoO<sub>2</sub>. The electric capacity may be increased by increasing the charge final voltage to higher than 4.2 V. However, with the increase of the charge capacity, the crystalline structure of LiCoO<sub>2</sub> decays so that the charge-discharge cycle life may be shortened, or the crystalline structure of LiCoO<sub>2</sub> may be destabilized. Accordingly, the thermal stability of the battery deteriorates.

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To solve such a problem, many attempts have been made to add a different metal element to  $\text{LiCoO}_2$  (cf. JP-A-4-171659, JP-A-3-201368, JP-A-7-176302 and JP-A-2001-167763).

In addition, attempts have been made to use a battery in a high-voltage range of 4.2 V or higher (cf. JP-A-2004-296098, JP-A-2001-176511 and JP-A-2002-270238). Also, attempts have been made to use a fluorine-containing organic solvent or a sulfur-containing organic solvent as a solvent of an electrolyte (cf. JP-A-8-37024, JP-A-10-112334, JP-A-10-112335, JP-A-2000-123880, JP-A-6-302336 and JP-A-11-162511).

In years to come, a secondary battery will be required to have a higher capacity and also better reliability including higher safety than the conventional batteries. In general, the battery capacity can be greatly improved by raising the content of an active material in electrodes or by increasing an electrode density, in particular, the density of a positive electrode mixture layer. However, according to such capacity-increasing measures, the safety of the battery gradually decreases.

Accordingly, in order to meet requirements for the increase of the electric capacity, it is highly desired to provide a battery which uses a material that generates a higher electromotive force (voltage range) than  ${\rm LiCoO_2}$  and has a stable crystalline structure capable of being stably and reversibly charged and discharged, and which further satisfies reliability such that the safety of the conventional batteries can be maintained and the battery does not expand when the density of the positive electrode mixture layer is increased.

When the discharge final voltage of a conventional battery comprising  $\text{LiCoO}_2$  as a positive electrode active material is made higher than 3.2 V, the battery cannot be completely discharged since the voltage in the final stage of the discharge significantly falls. Thus, an electric quantity efficiency of discharge relative to charging remarkably decreases. Since the complete discharge cannot be attained, the crystalline structure of  $\text{LiCoO}_2$  easily decays, and thus the charge-discharge cycle life is shortened. This phenomenon remarkably appears in the above-mentioned high voltage range.

Under a charging condition that the final voltage at full charging is set to 4.2 V or higher in the conventional battery, apart from shortening of the charge-discharge cycle life or the decrease of the thermal stability caused by the decay of the crystalline structure of the positive electrode active material, the electrolytic solution (a solvent) is oxidatively decomposed due to the increase of the active sites in the positive electrode active material, whereby a passivation film is formed on the surface of the positive electrode and thus the internal resistance of the battery increases so that the load characteristic may deteriorate.

# SUMMARY OF THE INVENTION

An object of the present invention is to provide a nonaqueous secondary battery having a high capacity, good chargedischarge cycle characteristics, and high reliability including atternal and a carbonaceous material such as graphite as a

Accordingly, the present invention provides a nonaqueous secondary battery comprising: a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electrolyte, wherein the positive electrode comprises, as active materials, at least two lithium-containing transition metal oxides having different average particle sizes, and at least the lithium-containing transition metal oxide having the smallest average particle size comprises at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn.

Furthermore, the present invention provides a nonaqueous secondary battery comprising: a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electrolyte, wherein the positive electrode comprises, as active materials, at least two lithium-containing 5 transition metal oxides having different average particle sizes, the lithium-containing transition metal oxides comprise at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn, and a ratio of a content (I) of said at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn in the transition metal oxide having the smallest average particle size to a content (II) of the same metal element as one with which the content (I) is measured, in the lithium-containing transition metal oxide other than one having the smallest average particle size is at least 1.5, where the contents (I) and (II) are measured by an inductively coupled plasma (ICP) atomic emission spectroscopy.

The "average particle size" of the lithium-containing transition metal oxides used herein means a 50% diameter value  $(d_{50})$ , that is, an median diameter, read from an integral fraction curve based on volumes, which is obtained by integrating the volumes of the particles from a smaller particle size measured by a MICROTRAC particle size analyzer (HRA 9320 25 available from NIKKISO Co., Ltd.).

The present invention also provides a method of using a nonaqueous secondary battery according to the present invention comprising the step of charging the battery so that a positive electrode voltage is in a range of 4.35 to 4.6 V with <sup>30</sup> reference to the potential of lithium when the battery is fully charged.

For example, in the method of using the nonaqueous secondary battery according to the present invention, when the nonaqueous secondary battery of the present invention comprises a graphite negative electrode, namely, a negative electrode containing graphite as a negative electrode active material, which has a voltage of 0.1 V with reference to the lithium potential when the battery is fully charged, charging the battery up to a battery voltage of 4.45 V or higher is regarded as 40 charging the battery so as to substantially attain a positive electrode voltage of 4.35 V or higher.

In the method of using the nonaqueous secondary battery according to the present invention, the term "fully charging (charged)" means charging under the following conditions: 45 the battery is charged at a constant current of 0.2 C up to a predetermined voltage and subsequently the battery is charged at a predetermined constant voltage, provided that the total time of the constant current charging and the constant voltage charging is set to 8 hours.

Consequently, the nonaqueous secondary battery of the present invention has a high capacity, good charge-discharge cycle characteristics and high reliability including high safety.

According to the using method of the present invention, the 55 nonaqueous secondary battery of the present invention can be used in applications which require larger power output.

# BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows an example of a particle size distribution curve of a positive electrode active material used in the non-aqueous secondary battery according to the present invention.

FIGS. 2A and 2B schematically show one example of the nonaqueous secondary battery of the present invention. FIG. 65 2A is a plan view thereof and FIG. 2B is a partial vertical section thereof.

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FIG. 3 shows a perspective view of the nonaqueous secondary battery illustrated in FIGS. 2A and 2B.

#### DETAILED DESCRIPTION OF THE INVENTION

The positive electrode used in the nonaqueous secondary battery of the present invention comprises, as the active material, the mixture of at least two lithium-containing transition metal oxides having different average particle sizes. When the lithium-containing transition metal oxide having a larger particle size and one having a smaller particle size are used in combination, gaps between the particles of one having the larger particle size are filled with the particles of one having the smaller particle size. Accordingly, the density of the positive electrode mixture layer is increased and, in turn, the capacity of the battery is increased.

The mixture of "at least two lithium-containing transition metal oxides having different average particle sizes" has three or more inflection points in a particle size distribution curve of the mixture. The particle size distribution curve may have two or more peaks or a shoulder in one or more peaks. In the case of such a particle size distribution curve, firstly, a conventional peak-separating method is applied to separate a distribution of particles having a larger particle size and that of particles having a smaller particle size. Subsequently, from the particle sizes and the integrated volume, the average particle size ( $d_{50}$ ) of each of the lithium-containing transition metal oxides and the mixing ratio between them can be calculated.

When the average particle size of the lithium-containing transition metal oxide having the largest average particle size (hereinafter referred to as "positive electrode active material (A)") is represented by A, and that of the lithium-containing transition metal oxide having the smallest average particle size (hereinafter referred to as "positive electrode active material (B)") is expressed by B, the ratio of B to A (i.e., B/A) is preferably from 0.15 to 0.6. When the average particle sizes of the two positive electrode active materials (A) and (B) have such a ratio B/A, the density of the positive electrode mixture layer can be easily increased.

The positive electrode active material (A) preferably has an average particle size of 5 µm or more, more preferably 8 µm or more, particularly preferably 11 µm or more. When the average particle size of the positive electrode active material 45 (A) is too small, the density of the positive electrode mixture layer may hardly be increased. When the average particle size is too large, the battery characteristic tends to decrease. Thus, the average particle size is preferably 25 µm or less, more preferably 20 µm or less, particularly preferably 18 µm or 50 less.

The positive electrode active material (B) preferably has an average particle size of 10  $\mu m$  or less, more preferably 7  $\mu m$  or less, particularly preferably 5  $\mu m$  or less. When the average particle size of the positive electrode active material (B) is too large, the positive electrode active material (B) does not easily fill the gaps between the particles of the lithium-containing transition metal oxide having a relatively large particle size in the positive electrode mixture layer, so that the density of this layer may hardly be increased. When the average particle size is too small, the volume of voids among the small particles increases so that the density of the positive electrode mixture layer may not be increased. Thus, the average particle size of the positive electrode active material (B) is preferably 2  $\mu$ m or more, more preferably 3  $\mu$ m or more, particularly preferably 4  $\mu$ m or more.

The positive electrode active materials according to the present invention may contain only two lithium-containing

transition metal oxides having different average particle sizes, for example, the positive electrode active materials (A) and (B) as descried above, while the positive electrode active materials may contain three or more, for example, three, four or five lithium-containing transition metal oxides having different average particle sizes, for example, the positive electrode active materials (A) and (B) and one or more lithium-containing transition metal oxides having an average particle size between those of the positive electrode active materials (A) and (B).

The content of the positive electrode active material (B) having the smallest average particle size in the lithium-containing transition metal oxides contained in the positive electrode is preferably 5% by weight or more, more preferably 10% by weight or more, particularly preferably 20% by 15 weight or more. When the positive electrode active material (B) is contained in an amount of the above-mentioned range, the gaps between the particles of the lithium-containing transition metal oxide having a relatively large particle size are easily filled therewith so that the density of the positive elec- 20 trode mixture layer is increased. When the content of the positive electrode active material (B) is too large, the density of the positive electrode mixture layer is hardly be increased. Thus, the content of the positive electrode active material (B) is preferably 60% by weight or less, more preferably 50% by 25 weight or less, particularly preferably 40% by weight or less.

Accordingly, when the lithium-containing transition metal oxides contained in the positive electrode are only the positive electrode active materials (A) and (B), the content of the positive electrode active material (A) is preferably 40% by weight or more, more preferably 50% by weight or more, particularly preferably 60% by weight or more of the oxides, while it is preferably 95% by weight or less, more preferably 90% by weight or less, particularly preferably 80% by weight or less.

Among the lithium-containing transition metal oxides contained in the positive electrode, the positive electrode active material (B) having the smallest average particle size has the above-mentioned average particle size. Such a lithium-containing transition metal oxide having a relatively small particle size has low stability, for example, in a state that the battery is charged at a high voltage, so that the oxide may damage the reliability including the safety of the battery.

Thus, the present invention uses, as at least the positive electrode active material (B), which is the lithium-containing 45 transition metal oxide having the smallest average particle size, a lithium-containing transition metal oxide comprising at least one metal element M<sup>2</sup> selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn, since the lithiumcontaining transition metal oxide comprising the metal ele- 50 ment M<sup>2</sup> has the improved stability, in particular, the stability in a state that the battery is charged at a high voltage, and thus the reliability including the safety of the battery can be improved. Furthermore, because of the improved stability of the lithium-containing transition metal oxide comprising the 55 metal element M<sup>2</sup>, the decay of the active material (B) is suppressed, when the charge-discharge cycles are repeated. Thus, the use of such a positive electrode active material (B) increases the charge-discharge cycle characteristics of the battery.

Apart from the positive electrode active material (B), the other lithium-containing transition metal oxide such as the positive electrode active material (A) and/or the lithium-containing transition metal oxide having the average particle size between those of the positive electrode active materials (A) and (B) may preferably comprise the metal element M<sup>2</sup>. When the lithium-containing transition metal oxide other

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than the positive electrode active material (B) comprises the metal element  $M^2$ , the reliability including the safety of the battery can be further improved, since such a lithium-containing transition metal oxide has the improved stability, in particular, the stability in a state that the battery is charged at a high voltage as described above.

The positive electrode active material (B) is preferably a lithium-containing transition metal oxide represented by the following formula (1):

$$\text{Li}_{x}\text{M}^{1}_{\nu}\text{M}^{2}_{z}\text{M}^{3}_{\nu}\text{O}_{2}$$
 (1)

wherein  $M^1$  represents at least one transition metal element selected from Co, Ni and Mn,  $M^2$  represents at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn,  $M^3$  represents an element other than Li,  $M^1$  and  $M^2$ , and x, y, z and v are numbers satisfying the following Equations respectively:  $0.97 \le x < 1.02$ ,  $0.8 \le y < 1.02$ ,  $0.002 \le z \le 0.05$ , and  $0 \le v \le 0.05$ . z is preferably at least 0.004, more preferably at least 0.006, while it is preferably less than 0.02, more preferably less than 0.01. When z is too small, the charge-discharge characteristics or the safety of the battery may not sufficiently be improved. When z is too large, the electric characteristics of the battery tend to deteriorate.

Each of the lithium-containing transition metal oxides other than the positive electrode active material (B) such as the positive electrode active material (A) is preferably a lithium-containing transition metal oxide represented by the following formula (2):

$$\operatorname{Li}_{a} \operatorname{M}^{1}_{b} \operatorname{M}^{2}_{c} \operatorname{M}^{3}_{d} \operatorname{O}_{2} \tag{2}$$

wherein  $M^1$ ,  $M^2$  and  $M^3$  are the same as defined in the formula (1), and a, b, c and d are numbers satisfying the following equations respectively:  $0.97 \le a < 1.02$ ,  $0.8 \le b < 1.02$ ,  $0 \le c \le 0.02$ , and 0 < d < 0.02.

M<sup>1</sup>, M<sup>2</sup> and M<sup>3</sup> are selected from the same elements as in the formula (1), but the elements selected or the constituting element ratios selected in the individual positive electrode active materials having different average particle sizes may differ from each other. For example, in the positive electrode active material (B), Mg, Ti and Al may be selected, while in the positive electrode active material (A), Mg and Ti may be selected. As explained in this example, however, among the elements M<sup>2</sup>, preferably at least one common element is selected, more preferably at least two common elements are selected, and particularly preferably at least three common elements are selected.

In the case of the positive electrode active material (A), "c" is preferably 0.0002 or more, more preferably 0.001 or more, and it is preferably less than 0.005, more preferably less than 0.0025, and "d" is preferably 0.0002 or more, more preferably 0.001 or more and it is preferably less than 0.005, more preferably less than 0.0025 for the following reason: the particle size of the positive electrode active material (A) is relatively large; thus, when the amount of  $M^2$  and the like added to the material (A) is relatively small, advantageous effects can be attained; but when the amount is too large, the electrical characteristics of the battery tends to decrease.

In each of the lithium-containing transition metal oxides used in the present invention, the transition metal element(s) thereof is/are preferably mainly Co and/or Ni. For example, the total amount of Co and Ni is preferably 50% by mole or more based on all the transition metal elements contained in the lithium-containing transition metal oxides.

Preferably, the proportion of Co in the lithium-containing transition metal oxide is higher, since the density of the positive electrode mixture layer can be made higher. In the formulae (1) and (2), the proportion of Co in the transition metal

element M<sup>1</sup> is preferably 30% by mole or more, more preferably 65% by mole or more, particularly preferably 95% by mole or more.

The values of x in the formula (1) and a in the formula (2)may vary as the battery is charged or discharged. Nevertheless, when the battery is an as-produced one, x and a are each preferably 0.97, more preferably 0.98 or more, particularly preferably 0.99 or more, while x and a are each preferably less than 1.02, more preferably 1.01 or less, particularly prefer-

The values of y in the formula (1) and b in the formula (2) are each preferably 0.98 or more, more preferably 0.98 or more, particularly preferably 0.99 or more, and they are each preferably less than 1.02, more preferably less than 1.01,  $_{15}$ particularly preferably less than 1.0.

Each of the positive electrode active material (B) represented by the formula (1), and the lithium-containing transition metal oxides other than the positive electrode active material (B) which are represented by the formula (2) pref- 20 erably contains Mg as the element M<sup>2</sup>, since the safety of the battery is more effectively improved. In addition, each of them comprises Mg and also at least one metal element M<sup>2</sup> selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn. In this case, the stability of those lithium-containing tran- 25 sition metal oxides is further improved in a state that the battery is charged at a high voltage.

In the positive electrode active material (B), the content of Mg is preferably at least 0.1% by mole, more preferably at least 0.15% by mole, particularly preferably at least 0.2% by mole, based on the amount of the metal element M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of Mg.

Herein the content of the metal element M<sup>2</sup> (% by mole) is based on the amount of the metal element M<sup>1</sup> (100% by mole).

When the positive electrode active material (B) contains at least one metal element selected from Ti, Zr, Ge and Nb, the total content thereof is at least 0.05% by mole, more preferably at least 0.08% by mole, particularly preferably at least of more effectively attaining the effects of the use of these metal elements. When the positive electrode active material (B) contains Al and/or Sn, the total content thereof is preferably 0.1% by mole or more, more preferably 0.15% by mole or more, particularly preferably 0.2% by mole or more based 45 on the content of M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of the use of these metal elements.

When the content of Mg is too large in the positive electrode active material (B), the load characteristic of the battery tends to decrease. Therefore, the content of Mg is preferably 50 less than 2% by mole, more preferably less than 1% by mole, particularly preferably 0.5% by mole, most preferably 0.3%

When the content of Ti, Zr, Ge, Nb, Al and/or Sn is too large in the positive electrode active material (B), the effect to 55 increase the capacity of the battery may not be sufficient. Thus, when the oxide contains Ti, Zr, Ge and/or Nb, the total content thereof is preferably less than 0.5% by mole, more preferably less than 0.25% by mole, particularly preferably less than 0.15% by mole based on the content of  $M^1$ . When the 60 oxide contains Al and/or Sn, the total amount thereof is preferably less than 1% by mole, more preferably less than 0.5% by mole, particularly preferably less than 0.3% by mole based on the content of M<sup>1</sup>.

In the positive electrode active material (A), the content of 65 Mg is preferably at least 0.01% by mole, more preferably at least 0.05% by mole, particularly preferably at least 0.07% by

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mole, based on the amount of the metal element M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of Mg.

When the positive electrode active material (A) contains at least one metal element selected from Ti, Zr, Ge and Nb, the total content thereof is at least 0.005% by mole, more preferably at least 0.008% by mole, particularly preferably at least 0.01% by mole, based on the content of M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of the use of these metal elements. When the positive electrode active material (A) contains Al and/or Sn, the total content thereof is preferably 0.01% by mole or more, more preferably 0.05% by mole or more, particularly preferably 0.07% by mole or more based on the content of M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of the use of these metal elements.

Also, when the content of Mg is too large in the positive electrode active material (A), the load characteristic of the battery tends to decrease. Therefore, the content of Mg is preferably less than 0.5% by mole, more preferably less than 0.2% by mole, particularly preferably 0.1° by mole.

Also, when the content of Ti, Zr, Ge, Nb, Al and/or Sn is too large in the positive electrode active material (A), the effect to increase the capacity of the battery may not be sufficient. Thus, when the oxide contains Ti, Zr, Ge and/or Nb, the total content thereof is preferably less than 0.3% by mole, more preferably less than 0.1% by mole, particularly preferably less than 0.05% by mole based on the content of M<sup>1</sup>. When the oxide contains Al and/or Sn, the total amount thereof is preferably less than 0.5% by mole, more preferably less than 0.2% by mole, particularly preferably less than 0.1% by mole based on the content of M<sup>1</sup>

Furthermore, when the lithium-containing transition metal oxide other than the positive electrode active materials (A) and (B) is used, the content of Mg in the other metal oxide is 35 preferably at least 0.01% by mole, more preferably at least 0.05% by mole, particularly preferably at least 0.07% by mole, based on the amount of the metal element M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of Mg.

When the lithium-containing transition metal oxide other 0.1% by mole, based on the content of M<sup>1</sup>, from the viewpoint 40 than the positive electrode active materials (A) and (B) contains at least one metal element selected from Ti, Zr, Ge and Nb, the total content thereof is at least 0.005% by mole, more preferably at least 0.008% by mole, particularly preferably at least 0.01% by mole, based on the content of M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of the use of these metal elements. When the positive electrode active material (A) contains Al and/or Sn, the total content thereof is preferably 0.01% by mole or more, more preferably 0.05% by mole or more, particularly preferably 0.07% by mole or more based on the content of M<sup>1</sup>, from the viewpoint of more effectively attaining the effects of the use of these metal elements.

> However, again in the lithium-containing transition metal oxide other than the positive electrode active materials (A) and (B), when the content of Mg is too large, the load characteristic of the battery tends to decrease. Therefore, the content of Mg is preferably less than 2% by mole, more preferably less than 1% by mole, particularly preferably 0.5% by mole, most preferably 0.3% by mole, each based on the content of M<sup>1</sup>.

> Again, in the lithium-containing transition metal oxide other than the positive electrode active materials (A) and (B), when the content of Ti, Zr, Ge, Nb, Al and/or Sn is too large in the positive electrode active material (A), the effect to increase the capacity of the battery may not be sufficient. Thus, when the oxide contains Ti, Zr, Ge and/or Nb, the total content thereof is preferably less than 0.5% by mole, more

preferably less than 0.25% by mole, particularly preferably less than 0.15% by mole based on the content of  $M^1$ . When the oxide contains Al and/or Sn, the total amount thereof is preferably less than 1% by mole, more preferably less than 0.5% by mole, particularly preferably less than 0.3% by mole based 5 on the content of  $M^1$ .

A method for including the metal element  $M^2$  in the positive electrode active material (B) or the other lithium-containing transition metal oxide(s) is not particularly limited. For example, the element  $M^2$  may be present on the particles of 10 the metal oxide, may be evenly present as a solid solution inside the metal oxides, or may be unevenly present inside the metal oxides with having a density distribution. Furthermore, the element  $M^2$  may form a compound which in turn forms a layer on the particle surfaces. Preferably, the element  $M^2$  is 15 evenly present as a solid solution.

In the formulae (1) and (2) representing the positive electrode active material (B) and the other lithium-containing transition metal oxide(s), respectively, the element M³ is an element other than Li, M¹ and M². The positive electrode 20 active material (B) and the other lithium-containing transition metal oxides may each contain the M³ in an amount such that the advantageous effects of the present invention are not impaired, or they may contain no M³.

Examples of the element M³ include alkali metals other than Li (e.g., Na, K and Rb), alkaline earth metals other than Mg (e.g., Be, Ca, Sr and Ba), Group IIIa metals (e.g., Sc, Y, La), Group IVa metals other than Ti and Zr (e.g., Hf), Group Va metals other than Nb (e.g., V and Ta), Group VIa metals (e.g., Cr, Mo and W), Group VIIb metals other than Mn (e.g., 30 Tc and Re), Group VIII metals other than Co and Ni (e.g., Fe, Ru, and Rh), Group Ib metals (e.g., Cu, Ag and Au), Group IIIb metals other than Zn and Al (e.g., B, Ca and In), Group IVb metals other than Sn and Pb (e.g., Si), P and Bi.

The metal element M² contributes to an improvement in the stability of the lithium-containing transition metal oxides. However, when the content thereof is too large, a function of storing and releasing Li ions is impaired so that the battery characteristics may be decreased. Since the positive electrode active material (B) having the smallest average particle size 40 has the particularly small particle size and decreased stability, it is preferable that the content of the element M², which is a stabilizing element, is somewhat high. In addition, since the positive electrode active material (B) has the small particle size and in turn the large surface area, it exhibits a high 45 activity. Thus, the presence of the element M² in the material (B) has less influence on the function of storing and releasing Li ions.

In contrast, the lithium-containing transition metal oxides having relatively large particle sizes, that is, the lithium-containing transition metal oxides other than the positive electrode active material (B), have better stability than the positive electrode active material (B). Therefore, the former metal oxides have less necessity to contain the element M² than the positive electrode active material (B). Furthermore, 55 their function of storing and releasing Li ions is easily impaired by the presence of the element M² since the materials have the smaller surface area and the lower activity than the positive electrode active material (B).

Accordingly, it is preferable that the content of the metal 60 element  $M^2$  in the positive electrode active material (B) is larger than that in the lithium-containing transition metal 0 oxide(s) other than the positive electrode active material (B).

That is, z in the formula (1) is preferably larger than c in the formula (2) (z>c). In particular, z is at least 1.5 times, more 65 preferably at least 2 times, particularly preferably at least 3 times larger than c. When z is much larger than c, the load

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characteristics of the battery tend to decrease. Thus, z is preferably less than 5 times as large as  $c^1$ , more preferably less than 4 times as large as c, particularly preferably less than 3.5 times as large as c.

When the three or more lithium-containing transition metal oxides having the different average particle sizes are contained in the positive electrode, there is no especial limitation on the relationship of the element M<sup>2</sup> content between the positive electrode active material (A) having the largest average particle size and the other lithium-containing transition metal oxides. Thus, the former may contain a larger amount of the element M<sup>2</sup> than the latter, and vise versa, or the element M<sup>2</sup> contents in the former and the latter may be the same. In a more preferable embodiment, a metal oxide having a smaller average particle size contains a larger amount of the element M<sup>2</sup>. In particular, when the three lithium-containing transition metal oxides having different average particle sizes are used, the element M<sup>2</sup> content in the positive electrode active material (B) having the smallest average particle size is largest, that in lithium-containing transition metal oxide having the average particle size between those of the active materials (A) and (B) is second largest, and that in the positive electrode active material (A) having the largest average particle size is smallest.

In the lithium-containing transition metal oxides which constitute the positive electrode active material according to the present invention, the oxides having different average particle sizes may have the same composition of elements, or different compositions of elements between them. When the lithium-containing transition metal oxides according to the present invention are the above-mentioned positive electrode active materials (A) and (B), the following combination may be used: a combination of the positive electrode active material (A) consisting of  $\text{LiCo}_{0.998}\text{Mg}_{0.0008}\text{Ti}_{0.0004}\text{Al}_{0.0008}\text{O}_2$ , and the positive electrode active material (B) consisting of  $\text{LiCo}_{0.334}\text{Ni}_{0.33}\text{Mn}_{0.33}\text{Mg}_{0.0024}\text{Ti}_{0.0012}\text{Al}_{0.0024}\text{O}_2$ .

The positive electrode active material comprising the lithium-containing transition metal oxides according to the present invention is formed through a certain synthesizing process and a certain battery producing process. For example, for the preparation of lithium-containing transition metal oxides which contain Co as the transition metal element M<sup>1</sup> and have different average particle sizes, firstly, a solution of an alkali such as NaOH is dropwise added to an acidic aqueous solution containing Co to precipitate Co(OH)<sub>2</sub>. In order to homogeneously precipitate Co(OH)<sub>2</sub>, Co may be coprecipitated with a different element, and then the coprecipitated material is calcined to obtain Co<sub>3</sub>O<sub>4</sub>. The particle size of the precipitates can be adjusted by controlling the period for forming the precipitates. The particle size of Co<sub>3</sub>O<sub>4</sub> after calcination is also controlled by the particle size of the precipitate at this time.

When the positive electrode active material is synthesized, conditions such as a mixing condition, calcination temperature, calcination atmosphere, calcination time, starting materials, and also battery fabrication conditions are suitably selected. With regard to the mixing condition in the synthesis of the positive electrode active material, preferably, for example, ethanol or water is added to the powdery starting materials, and then mixed in a planetary ball mill for 0.5 hour or longer. More preferably, ethanol and water are mixed at a volume ratio of 50:50, and the mixture is agitated in a planetary ball mill for 20 hours or longer. Through this mixing step, the powdery starting materials are sufficiently comminuted and mixed to prepare a homogeneous dispersion. The dispersion is dried with a spray drier or the like while keeping homogeneity. The calcination temperature is preferably from

 $750\, \rm to\, 1,050^{\circ}\, C.,$  more preferably from 950 to 1,030° C. The calcination atmosphere is preferably an air. The calcination time is preferably from 10 to 60 hours, more preferably from 20 to 40 hours.

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In the preparation of the positive electrode active material,  $^{5}$  Li $_{2}$ CO $_{3}$  is preferably used as a lithium source. As the sources of other metal such as Mg, Ti, Ge, Zr, Nb, Al and Sn, preferred are nitrates or hydroxides of these metals, or oxides thereof having a particle size of 1  $\mu$ m or less. It is preferable to use the coprecipitate of the hydroxides since the different elements  $^{10}$  are uniformly distributed in the active material.

The contents of the metal elements in the positive electrode active materials are measured by the ICP atomic emission spectroscopy or the like. The content of lithium can be measured by an atomic absorption analysis. In the state of a 15 positive electrode, it is difficult to separately measure the contents of the metal elements in each of the positive electrode active material having a larger particle size and one having a smaller particle size. Therefore, the contents or content ratios of the metal elements of the positive electrode 20 active material having different particle sizes may be measured with an electron prove microanalyzer using a mixture of positive electrode active materials having a known mixing ratio as a standard sample. Alternatively, the positive electrode is treated with a suitable solvent such as N-methyl-2- 25 pyrrolidone (NMP) to separate the active material particles from the positive electrode and settled out in the solvent, followed by washing and drying. Then, the particle size distribution of the recovered particles is measured and the peakseparation of the particle size distribution curve is carried out. 30 When the inclusion of two or more particles having different particle sized is confirmed, the particles are classified into a larger one and a smaller one, and the contents of the metal elements in each particle group are measured by the ICP atomic emission spectroscopy.

Herein, the contents of the metal elements in the positive electrode active material may be measured by the ICP atomic emission spectroscopy as follows: about 5 g of the active material is precisely weighed and charged in a 200 ml beaker. Then, 100 ml of aqua regia is added, and the mixture is 40 concentrated by heating to a liquid volume of about 20 to 25 ml. After cooling, the mixture is filtrated through a quantitative filter paper (No. SB available from Advantec MFS, Inc.) to separate the solids. The filtrate and washing liquid are charged in a 100 ml measuring flask and diluted to a specific 45 volume. Then, the contents of the metal elements in the solution are measured with a sequential type ICP analyzer (IPIS 1000 manufactured by Nippon Jarrel-Ash Co., Ltd.).

When the content (I) of at least one metal element selected from the group consisting of Mg, Ti, Zr, Ge, Nb, Al and Sn in 50 the transition metal oxide having the smallest average particle size and a content (II) of the same metal element as one with which the content (I) in the lithium-containing transition metal oxide other than one having the smallest average particle size are measured by the ICP atomic emission spectros- 55 copy described above, the ratio of the content (I) to the content (II) corresponds to the relationship between z in the formula (1) and c in the formula (2). The ratio of the content (I) to the content (II) is preferably at least 1.5, more preferably at least 2, particularly preferably at least 3. Since the load 60 characteristics of the battery tend to decrease when z is much larger than c, the ratio of the content (I) to the content (II) is preferably less than 5, more preferably less than 4, particularly preferably less than 3.5.

The positive electrode used in the present invention is 65 formed by, for example, a method described below. Firstly, the two or more lithium-containing transition metal oxides

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having different average particle sizes, for example, the positive electrode active materials (A) and (B), are mixed with each other at a predetermined weight ratio. If necessary, an electric conductive aid (e.g., graphite, carbon black, acetylene black, etc.) is added to the mixture. Furthermore, to the mixture, a binder (e.g., polyvinylidene fluoride, poly tetrafluoroethylene, etc.) is added to prepare a positive electrode mixture. A solvent is used to formulate this positive electrode mixture in the form of a paste. The binder may be mixed with the positive electrode active material and the like after the binder is dissolved in a solvent. In this way, the paste containing the positive electrode mixture is prepared. The resultant paste is applied to a positive electrode current collector made of an aluminum foil or the like, and then dried to form a positive electrode mixture layer. If necessary, the layer is pressed to obtain a positive electrode. However, the method for producing the positive electrode is not limited to the above-mentioned method, and may be any other method.

The positive electrode mixture layer according to the present invention may have a density of 3.5 g/cm³ or more, more preferably 3.6 g/cm³ or more, particularly preferably 3.8 g/cm³ or more. With such a density, the capacity of the battery can be increased. However, when the density of the positive electrode mixture layer is too high, the wettability with the nonaqueous electrolyte, which will be explained later, decreases. Thus, the density is preferably 4.6 g/cm³ or less, more preferably 4.4 g/cm³ or less, particularly preferably 4.2 g/cm³ or less.

Herein, the density of the positive electrode mixture layer may be obtained by the following measuring method: The positive electrode is cut to form a sample piece having a predetermined area, the sample piece is weighed with an electronic balance having a minimum scale of 1 mg, and then the weight of the current collector is subtracted from the 35 weight of the sample piece to calculate the weight of the positive electrode mixture layer. The total thickness of the positive electrode is measured at ten points with a micrometer having a minimum scale of 1 µm. Then, the thickness of the current collector is subtracted from the resultant individual thicknesses, and the thicknesses of the positive electrode mixture layer measured at ten points are averaged. From the averaged thicknesses of the positive electrode mixture layer and the surface area, the volume of the positive electrode mixture layer is calculated. Finally, the weight of the positive electrode mixture layer is divided by the volume thereof to obtain the density of the positive electrode mixture layer.

The thickness of the positive electrode mixture layer is preferably from 30 to 200  $\mu m$ , and the thickness of the current collector used in the positive electrode is preferably from 8 to 20  $\mu m$ .

In the positive electrode mixture layer, the content of the lithium-containing transition metal oxides as the active materials is preferably 96% by weight or more, more preferably 97% by weight or more, particularly preferably 97.5% by weight or more, while it is preferably 99% by weight or less, more preferably 98% by weight or less. The content of the binder in the positive electrode mixture layer is preferably 1% by weight or more, more preferably 1.3% by weight or more, particularly preferably 1.5% by weight or more, while it is preferably 4% by weight or less, more preferably 3% by weight or less, particularly preferably 2% by weight or less. The content of the electric conductive aid in the positive electrode mixture layer is preferably 1% by weight or more, more preferably 1.1% by weight or more, particularly preferably 1.2% by weight or more, while it is preferably 3% by weight or less, more preferably 2% by weight or less, particularly preferably 1.5% by weight or less.

When the content of the active material in the positive electrode mixture layer is too small, the capacity cannot be increased and also the density of the positive electrode mixture layer cannot be increased. When this content is too large, the resistance may increase or the formability of the positive 5 electrode may be impaired. When the binder content in the positive electrode mixture layer is too large, the capacity may hardly be increased. When this content is too small, the adhesion of the layer to the current collector decreases so that the powder may drop off from the electrode. Thus, the abovementioned preferable ranges are desirable. Furthermore, when the content of the electric conductive aid in the positive electrode mixture layer is too large, the density of the positive electrode mixture layer may not be made sufficiently high so that the capacity may hardly be increased. When this content 15 is too small, the sufficient electric conduction through the positive electrode mixture layer is not attained so that the charge-discharge cycle characteristic or the load characteristic of the battery may deteriorated.

It is essential for the nonaqueous secondary battery of the 20 present invention to have the positive electrode explained above, and thus there is no specific limitation on other elements or structure of the battery. The battery of the present invention may adopt various elements and structures, which are commonly adopted in the conventional nonaqueous secondary batteries in the state of art.

The negative electrode active material in the negative electrode may be any material that can be doped and de-doped with Li ions. Examples thereof are carbonaceous materials such as graphite, pyrolytic carbons, cokes, glassy carbons, 30 burned bodies of organic polymers, mesocarbon microbeads, carbon fibers and activated carbon. In addition, the following materials can also be used as the negative electrode active material: alloys of Si, Sn, In or the like, oxides of Si, Sn or the like that can be charged and discharged at a low voltage near 35 a voltage at which Li can be charged and discharged, and nitrides of Li and Co such as Li<sub>2.6</sub>Co<sub>0.4</sub>N. Graphite can be partially substituted with a metal, a metal oxide or the like that can be alloyed with Li. When graphite is used as the negative electrode active material, the voltage when the battery is fully 40 charged can be regarded as about 0.1 V with reference to the potential of lithium, and therefore the voltage of the positive electrode can be conveniently calculated as a voltage obtained by adding 0.1 V to the battery voltage. Consequently, the charge voltage of the positive electrode is easily controlled. 45

Preferably, graphite has such a form in that a lattice spacing  $d_{002}$  of the (002) planes is 0.338 nm or less, since the negative electrode or a negative electrode mixture layer, which will be explained later, has a higher density as the crystallinity is higher. However, when the lattice spacing  $d_{002}$  is too large, the 50 high density negative electrode may decrease the discharge characteristic or the load characteristic of the battery. Thus, the lattice spacing  $d_{002}$  is preferably 0.335 nm or more, more preferably 0.3355 nm or more.

The crystal size of the graphite in the c axis direction (Lc) 55 is preferably 70 nm or more, more preferably 80 nm or more, particularly preferably 90 nm or more. As the Lc is larger, the charging curve becomes flat so that the voltage of the positive electrode is easily controlled and also the capacity can be made large. When the Lc is too large, the battery capacity 60 tends to decrease with the high-density negative electrode. Thus, the Lc is preferably less than 200 nm.

Furthermore, the specific surface area of the graphite is preferably  $0.5~\text{m}^2/\text{g}$  or more, more preferably  $1~\text{m}^2/\text{g}$  or more, particularly preferably  $2~\text{m}^2/\text{g}$  or more, while it is preferably  $6~\text{m}^2/\text{g}$  or less, more preferably  $5~\text{m}^2/\text{g}$  or less. Unless the specific surface area of the graphite is somewhat large, the

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characteristics tend to decrease. When the specific surface area is too large, the graphite easily reacts with the electrolyte and such a reaction may have influences on the properties of the battery.

The graphite used in the negative electrode is preferably made of natural graphite. More preferred is a mixture of two or more graphite materials having different surface crystal-linity to achieve the high density of the negative electrode. Since natural graphite is inexpensive and achieves a high capacity, the negative electrode with a high cost performance can be produced. Usually, when natural graphite is used, the battery capacity is easily decreased as the density of the negative electrode is increased. However, the decrease in the battery capacity can be suppressed by mixing the natural graphite with a graphite having a reduced surface crystallinity by a surface treatment.

The surface crystallinity of specific graphite can be determined by the Raman spectrum analysis. When the R value of the Raman spectrum ( $R=I_{1350}/I_{1580}$ , that is, the ratio of the Raman intensity around 1350 cm<sup>-1</sup> to that around 1580 cm<sup>-1</sup>) is 0.01 or more, where the Raman spectrum is measured with graphite which has been excited with an argon laser having a wavelength of 514.5 nm, the surface crystallinity of the specific graphite is slightly lower than that of natural graphite. Thus, with the graphite having a surface crystallinity decreased by the surface treatment, the R value is preferably 0.01 or more, more preferably 0.1 or more, while it is preferably 0.5 or less, more preferably 0.3 or less. The content of the graphite having a surface crystallinity decreased by surface treatment is preferably 100% by weight of the whole graphite in order to increase the density of the negative electrode. However, in order to prevent the decrease of the battery capacity, the content of such graphite is preferably 50% by weight or more, more preferably 70% by weight or more, particularly preferably 85% by weight or more of the whole graphite.

When the average particle size of the graphite is too small, an irreversible capacity increases. Thus, the average particle size of the graphite is preferably 5  $\mu m$  or more, more preferably 12  $\mu m$  or more, particularly preferably 18  $\mu m$  or more. From the viewpoint of the increase of the capacity of the negative electrode, the average particle size of the graphite is 30  $\mu m$  or less, more preferably 25  $\mu m$  or less, particularly preferably 20  $\mu m$  or less.

The negative electrode may be produced by the following method, for example: The negative electrode active material and an optional a binder and/or other additives are mixed to prepare a negative electrode mixture, and the mixture is dispersed in a solvent to prepare a paste. Preferably, the binder is dissolved in a solvent prior to mixing with the negative electrode active material, and then mixed with the negative electrode active material and so on. The paste containing the negative electrode mixture is applied to a negative electrode current collector made of a copper foil or the like, and then dried to form a negative electrode mixture layer. The layer is pressed to obtain a negative electrode. However, the method for producing the negative electrode is not limited to the above-mentioned method, and may be any other method.

The density of the negative electrode mixture layer after pressing is preferably 1.70 g/cm³ or more, more preferably 1.75 g/cm³ or more. Based on the theoretical density of graphite, the upper limit of the density of the negative electrode mixture layer formed using graphite is 2.1 to 2.2 g/cm³. The density of the negative electrode mixture layer is preferably 2.0 g/cm³ or less, more preferably 1.9 g/cm³ or less from the viewpoint of the affinity with the nonaqueous electrolyte. It is

preferable to press the negative electrode plural times since the negative electrode can be uniformly pressed.

The binder used in the negative electrode is not particularly limited. For the increase of the content of the active material to increase the capacity, the amount of the binder is preferably 5 made as small as possible. To this end, the binder is preferably a mixture of an aqueous resin which can be dissolved or dispersed, and a rubbery polymer, since the use of only a small amount of the aqueous resin can contribute to the dispersion of the graphite and thus prevents the delamination of 10 the negative electrode mixture layer from the current collector caused by the expansion and contraction of the electrode in the charge-discharge cycles.

Examples of the aqueous resins include cellulose resins such as carboxymethylcellulose, and hydroxypropylcellu- 15 lose, and polyvinylpyrrolidone, polyepichlorohydrin, polyvinylpyridine, polyvinyl alcohol, polyether resins such as polyethylene oxide and polyethylene glycol, etc. Examples of the rubbery polymers include latex, butyl rubber, fluororubber, styrene-butadiene rubber, nitrile-butadiene copolymer rub- 20 ber, ethylene-propylene-diene copolymer (EPDM), polybutadiene, etc. From the viewpoint of the dispersibility of the graphite particles and the prevention of delamination of the layer, it is preferable to use a cellulose ether compound such as carboxymethylcellulose together with a butadiene copoly- 25 mer rubber such as a styrene-butadiene rubber. It is particularly preferable to use carboxymethylcellulose together with a butadiene copolymer rubber such as a styrene-butadiene copolymer rubber or a nitrile-butadiene-copolymer rubber. The cellulose ether compound such as carboxymethylcellu- 30 lose mainly has a thickening effect on the paste containing the negative electrode mixture, while the rubbery polymer such as the styrene-butadiene copolymer rubber has a binding effect on the negative electrode mixture. When the cellulose ether compound such as carboxymethylcellulose and the rub- 35 bery polymer such as the styrene-butadiene copolymer rubber are used in combination, the weight of the former to the latter is preferably from 1:1 to 1:15.

The thickness of the negative electrode mixture layer is preferably from 40 to 200  $\mu$ m. The thickness of the current 40 collector used in the negative electrode is preferably from 5 to 30  $\mu$ m.

In the negative electrode mixture layer, the content of the binder or binders is preferably 1.5% by weight or more, more preferably 1.8% by weight or more, particularly preferably 45 2.0% by weight or more of the layer, while it is preferably less than 5% by weight, less than 3% by weight, less than 2.5% by weight. When the amount of the binder in the negative electrode mixture layer is too large, the discharge capacity of the battery may decrease. When the amount is too small, the 50 adhesion between the particles decreases. The content of the negative electrode active material in the negative electrode mixture layer is preferably more than 95% by weight and 98.5% by weight or less.

In the nonaqueous secondary battery of the present invention, the nonaqueous electrolyte is preferably a nonaqueous solvent-base electrolytic solution comprising an electrolyte salt such as a lithium salt dissolved in a nonaqueous solvent such as an organic solvent, from the viewpoint of electric characteristics or handling easiness. A polymer electrolyte or 60 a gel electrolyte may be used without any problem.

The solvent in the nonaqueous electrolytic solution is not particularly limited, and examples thereof include acyclic esters such as dimethyl carbonate, diethyl carbonate, ethylmethyl carbonate, and methyl propyl carbonate; cyclic esters having a high dielectric constant, such as ethylene carbonate, propylene carbonate, butylene carbonate, and vinylene car-

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bonate; and mixed solvents comprising an acyclic ester and a cyclic ester. Mixed solvents each comprising an acyclic ester as a main solvent and a cyclic ester are particularly suitable.

Apart form the esters exemplified above, the following solvents may also be used: acyclic phosphoric acid triesters such as trimethyl phosphate; ethers such as 1,2-dimethoxyethane, 1,3-dioxolane, tetrahydrofuran, 2-methyl-tetrahydrofuran and diethyl ether; nitrites and dinitriles; isocyanates; and halogen-containing solvents. Furthermore, amine or imide organic solvents may be used.

Furthermore, the nonaqueous electrolytic solution may contain a fluorine-containing organic solvent, which can improve the stability of the battery since it forms a surface protective film on the positive electrode active materials during charging. More preferably, a fluorine-containing organic solvent having a carbon-carbon double bond may be used in combination with other usual fluorine-containing organic solvent. Specific examples of the fluorine-containing organic solvent include HFE7100 (C<sub>4</sub>F<sub>9</sub>OCH<sub>3</sub>), F-DPC [C<sub>2</sub>F<sub>5</sub>CH<sub>2</sub>O  $(C=O)OCH_2C_2F_5$ , F-DEC  $[CF_3CH_2O(C=O)OCH_2CF_3]$ , fluorobenzene, fluoroethylene carbonate [(—OCH<sub>2</sub>— CHFO—(C=O), (—OCHF—CHFO—)C=O, etc.], and the like. The amount of such a solvent added is preferably at least 0.1% by weight, more preferably at least 2% by weight, particularly preferably at least 5% by weight, while it is preferably 30% by weight or less, more preferably 20% by weight or less, particularly preferably 15% by weight or less, based on the whole weight of the electrolyte. When the amount of such a solvent is too large, the electric characteristics of the battery tend to decrease. When it is too small, any good protective film may not be formed.

The nonaqueous electrolyte may contain a sulfur-containing organic solvent, which can improve the stability of the battery since it forms a surface protective film on the positive electrode active materials during charging. The sulfur-containing organic solvent is preferably an organic compound having —O—SO<sub>2</sub>— bond, more preferably one having a -O—SO<sub>2</sub>—O— bond. Specific examples of such a solvent include sulfone, methylsulfolane, methylethylene sulfate, 1,3-propanesulfone, diethyl sulfate, butyl sulfate, butylsulfone, diphenyldisulfide, etc. The amount of such a solvent is preferably at least 0.1% by weight, more preferably at least 1% by weight, particularly preferably at least 2% by weight, while it is preferably 10% by weight or less, more preferably 5% by weight or less, particularly preferably 3% by weight or less, based on the whole weight of the electrolyte. When the amount of such a solvent is too large, the electric characteristics of the battery tend to decrease. When it is too small, any good protective film may not be formed.

Furthermore, the nonaqueous electrolytic solution may preferably contain a nonionic aromatic compound. Specific examples thereof include aromatic compounds having an alkyl group bonded to an aromatic ring (e.g., cyclohexylbenzene, isopropylbenzene, tert-butylbenzene, tert-amylbenzene, octylbenzene, toluene and xylene); aromatic compounds having a halogen group bonded to an aromatic ring (e.g., fluorobenzene, difluorobenzene, trifluorobenzene and chlorobenzene); aromatic compounds having an alkoxy group bonded to an aromatic ring (e.g., anisole, fluoroanisole, dimethoxybenzene and diethoxybenzene); aromatic carboxylic acid esters such as phthalic acid esters (e.g., dibutyl phthalate and di-2-ethylhexyl phthalate) and benzoic acid esters; carbonic acid esters having a phenyl group (e.g., methylphenyl carbonate, butylphenyl carbonate and diphenyl carbonate); phenyl propionate; and biphenyl. Among them, the

compounds having an alkyl group bonded to an aromatic ring (alkaryl compounds) are preferred, and cyclohexylbenzene is particularly preferred.

The aromatic compounds exemplified above can form a film on the surface of the active material in the positive 5 electrode or the negative electrode in the battery. These aromatic compounds may be used alone, while more advantageous effects can be attained by the use of two or more of the aromatic compounds together. Particularly advantageous effects can be attained on the improvement of the safety of the 10 battery by the use of the alkaryl compound together with an aromatic compound, which can be oxidized at a lower voltage than the alkaryl compound, such as biphenyl.

The method for adding the aromatic compound in the non-aqueous electrolytic solution is not particularly limited. In 15 general, the aromatic compound is added to the nonaqueous electrolytic solution prior to the fabrication of the battery.

The content of the aromatic compound in the nonaqueous electrolytic solution is preferably 4% by weight or more from the viewpoint of the safety, and it is preferably 10% by weight 20 or less from the viewpoint of the load characteristic. When two or more aromatic compounds are used together, the total amount thereof is within the above-mentioned range. When the alkaryl(alkylaryl) compound and the aromatic compound which can be oxidized at a lower voltage that the alkaryl 25 compound are used in combination, the content of the alkaryl compound in the nonaqueous electrolytic solution is preferably 0.5% by weight or more, more preferably 2% by weight or more, while it is preferably 8% by weight or less, more preferably 5% by weight or less. The content of the aromatic 30 compound that can be oxidized at a lower voltage than the alkaryl compound in the nonaqueous electrolytic solution is preferably 0.1% by weight or more, more preferably 0.2% by weight or more, while it is preferably 1% by weight or less, more preferably 0.5% by weight or less.

Furthermore, a surface protecting coating can be formed on the surface of the positive electrode active material in the step of initial charging of the battery, when the nonaqueous electrolytic solution contains at least one compound selected from the group consisting of halogen-containing organic sol- 40 vents (e.g., halogen-containing carbonates), fluorine-containing organic lithium salts, phosphorus-containing organic solvents, silicon-containing organic solvents, nitrogen-containing organic solvents, etc. Specific examples thereof include F-DPC [C<sub>2</sub>F<sub>5</sub>CH<sub>2</sub>O(C=O)OCH<sub>2</sub>C<sub>2</sub>F<sub>5</sub>], F-DEC 45 [CF<sub>3</sub>CH<sub>2</sub>O(C=O)OCH<sub>2</sub>CF<sub>3</sub>], HFE7100 (C<sub>4</sub>F<sub>9</sub>OCH<sub>3</sub>), butyl sulfate  $(C_4H_9OSO_2OC_4H_9)$ , methylethylene sulfate  $[(-OCH(CH_3)CH_2O-)SO_2],$ butyl (C<sub>4</sub>H<sub>9</sub>SO<sub>2</sub>C<sub>4</sub>H<sub>9</sub>), a polymer imide salt ([—N(Li)SO<sub>2</sub>OCH<sub>2</sub>  $(CF_2)_4$ — $CH_2OSO_2$ —]<sub>n</sub> wherein n is from 2 to 100), 50  $(C_2F_5SO_2)_2NLi$ , and  $[(CF_3)_2CHOSO_2]_2NLi$ .

Such additives may be used alone. It is particularly preferable to use a fluorine-containing organic solvent together with a fluorine-containing organic lithium salt. The amount of such an additive added is preferably 0.1% by weight or more, 55 more preferably 2% by weight or more, particularly preferably 5% by weight or more, while it is preferably 30% by weight or less, more preferably 10% by weight or less, each based on the whole weight of the nonaqueous electrolytic solution. When the amount of the additive is too large, the 60 electric characteristics of the battery may deteriorate. When the amount is too small, a good coating may hardly be formed.

When the battery comprising the nonaqueous electrolytic solution containing the above-mentioned additive(s) is charged, particularly at a high voltage, a surface protecting coating that contains fluorine or sulfur atoms is formed on the positive electrode active material surface. This surface pro-

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tecting coating may contain either fluorine atoms or sulfur atoms. Preferably, the coating contains both of fluorine atoms and sulfur atoms.

The amount of the sulfur atoms in the surface protecting coating formed on the positive electrode active material surface is preferably 0.5 atomic % or more, more preferably 1 atomic % or more, particularly preferably 3 atomic % or more. However, when the amount of the sulfur atoms in the positive electrode active material surface is too large, the discharge characteristic of the battery tends to decrease. Thus, the amount is preferably 20 atomic % or less, more preferably 10 atomic % or less, particularly preferably 6 atomic % or less.

The amount of the fluorine atoms in the surface protecting coating formed on the positive electrode active material surface is preferably 15 atomic % or more, more preferably 20 atomic % or more, particularly preferably 25 atomic % or more. However, when the amount of the fluorine atoms in the positive electrode active material surface is too large, the discharge characteristic of the battery tends to fall. Thus, the amount of the fluorine atoms is preferably 50 atomic % or less, more preferably 40 atomic % or less, particularly preferably 30 atomic % or less. The surface protecting coating, which contains the fluorine atoms and/or the sulfur atoms, in the positive electrode active material may not be formed by charging the battery as described above, but the positive electrode (battery) may be formed by the use of the positive electrode active material, that is, the lithium-containing transition metal oxides, which has such a surface protecting coating already formed.

In order to improve the charge-discharge cycle characteristic of the battery, preferably, the nonaqueous electrolytic solution contains at least one carbonate compound selected from the group consisting of vinylene carbonates, such as 35 (—OCH=CHO—)C=O, (—OCH=C(CH<sub>3</sub>)O—)C=O and (—OC(CH<sub>3</sub>)—C(CH<sub>3</sub>)O—)C—O, and derivatives thereof; and fluorine-substituted ethylene carbonates, such as -OCH<sub>2</sub>--CHFO--)C=O and (--OCHF--CHFO--) C=O. The addition amount thereof is preferably 0.1% by weight or more, more preferably 0.5% by weight or more, particularly preferably 2% by weight or more based on the whole weight of the nonaqueous electrolytic solution. When the addition amount thereof is too large, the load characteristic of the battery tends to decrease. Thus, the addition amount is preferably 10% by weight or less, more preferably 5% by weight or less, particularly preferably 3% by weight or less based on the whole weight of the nonaqueous electrolytic

Examples of the electrolyte salt to be dissolved in the solvent during the preparation of the nonaqueous electrolytic solution include LiClO<sub>4</sub>, LiPF<sub>6</sub>, LiBF<sub>4</sub>, LiAsF<sub>6</sub>, LiSbF<sub>6</sub>, LiCF<sub>3</sub>SO<sub>3</sub>, LiC<sub>4</sub>F<sub>9</sub>SO<sub>3</sub>, LiCF<sub>3</sub>CO<sub>2</sub>, Li<sub>2</sub>C<sub>2</sub>F<sub>4</sub>(SO<sub>3</sub>)<sub>2</sub>, LiN  $(\mathsf{RfSO}_2)(\mathsf{RfSO}_2), \mathsf{LiC}(\mathsf{RfSO}_2)_3, \mathsf{LiC}_n \mathsf{F}_{2n+1} \mathsf{SO}_3 \, \mathsf{wherein} \, \mathsf{n} {\geq} 2,$ and LiN(RfOSO<sub>2</sub>)<sub>2</sub> wherein Rf and Rf each represent a fluoroalkyl group. They may be used alone or in combination of two or more thereof. Among these electrolyte salts, particularly preferred are fluorine-containing organic lithium salts having 2 or more carbon atoms, since such lithium salts have a large anionic property and further ion separation easily occurs so that the salts are easily dissolved in the abovementioned solvents. The concentration of the electrolyte salt in the nonaqueous electrolytic solution is not particularly limited, and it is preferably 0.3 mol/L or more, more preferably 0.4 mol/L or more, while it is preferably 1.7 mol/L or less, more preferably 1.5 mol/L or less.

In the present invention, the nonaqueous electrolyte may be a gel-form polymer electrolyte besides the nonaqueous elec-

trolytic solution described above. The gel-form polymer electrolyte corresponds to a product obtained by the gelation of the nonaqueous electrolytic solution with a gelling agent. For the gelation of the nonaqueous electrolytic solution, the following gelling agents may be used: a linear polymer such as 5 polyvinylidene fluoride, polyethylene oxide or polyacrylonitrile, or a copolymer thereof; or a polyfunctional monomer which can be polymerized by irradiation with actinic rays such as ultraviolet rays or electron beams (e.g., an acrylate having 4 or more functionalities such as pentaerythritol tet- 10 raacrylate, ditrimethylolpropane tetraacrylate, ethoxylated pentaerythritol tetraacrylate, dipentaerythritol hydroxypentaacrylate, or dipentaerythritol hexaacrylate; or a methacrylate having 4 or more functionalities, which are analogous to the above acrylates. In the case of the monomer, the monomer itself does not cause the gelling of the electrolytic solution, but the polymer formed from the monomer acts as a gelling agent.

When a polyfunctional monomer is used to gel the electrolytic solution as described above, a polymerization initiator 20 may optionally be used. Examples of the polymerization initiator include benzoyls, benzoin alkyl ethers, benzophenones, benzoylphenylphosphine oxides, acetophenones, thioxanthones and anthraquinones. As a sensitizer for the polymerization initiator, an alkylamine or an aminoester may 25 be used.

In the present invention, the nonaqueous electrolyte may be a solid electrolyte besides the nonaqueous electrolytic solution or the gel-form polymer electrolyte. The solid electrolyte may be an inorganic solid electrolyte or an organic solid 30 electrolyte.

In the present invention, a separator used in the present invention preferably has a thickness of 5 µm or more, more preferably 10 µm or more, particularly preferably 12 µm or more, while it is preferably less than 25 µm, more preferably 35 less than 20 μm, particularly preferably less than 18 μm, from the viewpoint of imparting the directionality of the tensile strength to the separator, keeping good insulating properties and reducing the thermal shrinkage of the separator. The gas permeability of the separator is preferably 500 second/100-40 mL or less, more preferably 300 second/100-mL or less, particularly preferably 120 second/100-mL or less. As the gas permeability of the separator is smaller, the load characteristic is made better but an inside short-circuit is more easily caused. Thus, the gas permeability is preferably 50 second/ 45 100-mL or more. Here, a gas permeability is measured according to JIS P8117. As the thermal shrinkage factor of the separator in the transverse direction (TD) is smaller, an inside short-circuit is less easily caused when the temperature of the battery rises. Thus, the thermal shrinkage factor in TD of the 50 separator is as small as possible. The thermal shrinkage factor in TD is preferably 10% or less, more preferably 5% or less. In order to restrain the thermal shrinkage of the separator, it is preferable to thermally treat the separator in advance at a temperature of about 100 to 125° C. The separator having 55 such a thermal shrinkage factor is preferably combined with the positive electrode materials according to the present invention to fabricate a battery, since the behaviors of the battery at high temperature become stable.

The thermal shrinkage factor in TD of the separator means  $^{60}$  the shrinkage factor of a portion thereof that most largely shrinks in TD when the separator having a size of 30 mm square is allowed to stand at  $^{105}$ ° C. for 8 hours.

With regard to the strength of the separator, a tensile strength in the machine direction (MD) is preferably  $6.8 \times 10^7$  N/m $^2$  or more, more preferably  $9.8 \times 10^7$  N/m or more. The tensile strength in TD is preferably smaller than that in MD.

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The ratio of the tensile strength in TD to that in MD (tensile strength in TD/tensile strength in MD) is preferably 0.95 or less, more preferably 0.9 or less, while it is preferably 0.1 or more. The transverse direction means a direction perpendicular to the direction in which the film resin for the production of the separator is wound up, that is, the machine direction.

The puncture strength of the separator is preferably 2.0 N or more, more preferably 2.5 N or more. As this value is higher, the battery is less easily short-circuited. Usually, however, the upper limit thereof is substantially determined by the material of the separator. In the case of a separator made of polyethylene, the upper limit of the puncture strength is about 10 N. Here, a puncture strength is measured by cutting a sample piece of 50 mm×50 mm from a separator, clamping the sample piece with jigs at the edges of 5 mm, puncturing the sample piece with a needle having a tip end with a radius of 0.5 mm at a rate of 2 mm/sec., and measuring a maximum load before the puncture of the sample piece as a puncture strength.

When a conventional nonaqueous secondary battery is charged at a high positive electrode voltage of 4.35 V or higher with reference to the potential of lithium and is discharged to a final voltage higher than 3.2 V, the crystalline structure of the positive electrode active material decays to decrease the capacity or to induce heating of the battery due to the deterioration of the thermal stability. Thus, the battery may not be practically used. When a positive electrode active material to which a different element such as Mg or Ti is added is used, the decrease of the safety or of the capacity over charge-discharge cycles can be suppressed, but the degree of suppression is not sufficient. Moreover, the filling of the positive electrode is insufficient so that the battery easily expands.

In contrast, the battery of the present invention having the structure explained above is a nonaqueous secondary battery which improves the capacity, the charge-discharge cycle characteristic, the safety and the suppression of expansion of the battery. These advantageous effects can be attained at a usual charging volt (a battery voltage of 4.2 V). Furthermore, when the positive electrode is charged up to a high voltage of 4.35 V with reference to the potential of lithium (i.e., a battery voltage of 4.25 V) and then the discharge of the battery is terminated at a high voltage, that is, a battery voltage of 3.2 V or higher, the crystalline structures of the positive electrode active materials are very stable so that the decrease of the capacity or thermal stability is prevented.

Moreover, the positive electrode active material of any conventional nonaqueous secondary battery generates a low average voltage. Therefore, when a charge-discharge cycle test is repeated under a condition that the discharge final voltage of a unit cell is 4.35 V or higher with reference to the potential of lithium, the positive electrode is doped or dedoped with a large amount of Li ions. This situation is analogous to a case where the battery is subjected to a chargedischarge cycle test under an overcharge condition. Under such a severe condition, any conventional positive electrode active material cannot maintain its crystalline structure so as to cause disadvantages such that the thermal stability declines or the charge-discharge cycle life is shortened. To the contrary, the use of the positive electrode active materials according to the battery of the present invention can overcome such disadvantages of the conventional positive electrode active material. Thus, the present invention provides a nonaqueous secondary battery which can be reversibly charged and discharged even at a high voltage, such as a voltage of 4.35 to 4.6 V with reference to the potential of lithium.

The nonaqueous secondary battery of the present invention has characteristics including a high voltage, a high capacity and a high safety. By making use of such characteristics, the nonaqueous secondary battery of the present invention can be used as a power source of a notebook personal computer, a stylus-operated personal computer, a pocket personal computer, a notebook word processor, a pocket word processor, an electronic book player, a cellular phone, a codeless handset, a pager, a portable terminal, a portable copier, an electrical notebook, an electronic Calculator, a liquid crystal television set, an electric shaver, an electric power tool, an electronic translating machine, an automobile telephone, a transceiver, a voice input device, a memory card, a backup power source, a tape recorder, a radio, a headphone stereo, a handy printer, a handy cleaner, a portable CD player, a video movie, a navigation system, a refrigerator, an air conditioner, a television, a stereo, a water heater, a microwave oven, a dishwasher, a washing machine, a drying machine, a game equipment, a lighting equipment, a toy, a sensor equipment, a load conditioner, a medical machine, an automobile, an electric vehicle, 20 a golf cart, an electrically-powered cart, a security system, a power storing system, or the like. The battery can be used not only for the consumer applications but also for aerospace applications. The capacity-increasing effect of the present invention is enhanced, in particular, in small-sized portable 25 devices. Thus, the battery of the present invention is used in a portable device desirably having a weight of 3 kg or less, more desirably 1 kg or less. The lower limit of the weight of the portable device is not particularly limited. However, the lower limit is desirably a value equal to the weight of the battery, for example, 10 g or more in order to attain the advantageous effects to some degree.

# **EXAMPLES**

The present invention will be described in detail with reference to the following Examples; however, the Examples do not limit the scope of the present invention. Thus, modifications of the examples are encompassed by the scope of the present invention as long as the modifications do not depart 40 from the subject matter of the present invention, which has been described above or will be described hereinafter.

#### Example 1

# Production of Positive Electrode

The lithium-containing positive electrode materials,  $LiCo_{0.998}Mg_{0.0008}Ti_{0.0004}Al_{0.0008}O_2$  (average particle size: 12 µm) as a positive electrode active material (A), and 50  $LiCo_{0.994}Mg_{0.0024}Ti_{0.0012}Al_{0.0024}O_{2} \ (average\ particle\ size:5$ μm) as a positive electrode active material (B) at a weight ratio of 65:35 were mixed. Then, 97.3 parts by weight of the mixture and 1.5 parts by weight of a carbonaceous material as an electric conductive aid were charged in a volumetric feeder 55 as a device for supplying powder. An amount of a 10 wt. % solution of polyvinylidene fluoride (PVDF) in N-methyl-2pyrrolidone (NMP) to be supplied to the feeder was adjusted to control a solid content in the mixture constantly at 94% by weight during kneading. While the amount of the mixed 60 materials supplied in a unit time was controlled to a predetermined amount, the materials were supplied in a biaxial kneading extruder and then kneaded. In this way, a paste containing the positive electrode mixture was prepared.

Separately, the positive electrode active materials (A) and 65 (B) were dissolved in aqua regia and the ratio of the elements contained in the materials (A) and (B) was measured by the

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ICP atomic emission spectroscopy, the results of which confirmed that they had the above elementary compositions.

The resultant paste was charged in a planetary mixer, and then a 10 wt. % solution of PVDF in NMP, and NMP were added to dilute the paste, thereby adjusting the viscosity of the paste at a level sufficient for application. This diluted paste containing the positive electrode active material mixture was passed through a 70-mesh net to remove large substances. Thereafter, the paste was uniformly applied to both surfaces of a positive electrode current collector made of an aluminum foil with a thickness of 15 μm, and then dried to form filmform positive electrode mixture layers. In the dried positive electrode mixture layers, the weight ratio of the positive electrode active material/the electric conduction aiding agent/ PVDF was 97.3:1.5:1.2. Thereafter, the resultant sheet was pressed and cut out in a predetermined size. To the cut piece, a lead member made of aluminum was welded to form a sheet-form positive electrode. The density of the pressed positive electrode mixture layers (the density of the positive electrode) was 3.8 g/cm<sup>3</sup>. The thickness of the positive electrode mixture layers (the total thickness of the layers on both the surfaces, i.e., the thickness obtained by subtracting the thickness of the aluminum foil layer of the positive electrode current collector from the total thickness of the positive electrode) was 135 µm.

The particle size distribution of the mixture of the positive electrode active materials (A) and (B) was measured by a MICROTRAC particle size analyzer (HRA 9320 available from NIKKISO Co., Ltd.). The result is shown in the graph of FIG. 1, which confirm that the mixture contained two kinds of the particles having an average particle size of about 5  $\mu$ m and 12  $\mu$ m, respectively.

In the positive electrode active material (A), the amount of Mg was 0.08% by mole, that of Ti was 0.04% by mole, and that of Al was 0.08% by mole, each based on the amount of Co. An electron prove X-ray microanalyzer (EMPA 1600 manufactured by Shimadzu Corporation) was used to measure the concentration of the metal element M² in cross sections of the particles. As a result, no difference in the concentration of each of Mg, Ti and Al was observed between the surface portion and the core portion.

In the positive electrode active material (B), the amount of Mg was 0.24% by mole, that of Ti was 0.12% by mole, and that of Al was 0.24% by mole, each based on the amount of Co. The concentration of the metal element M² in the cross sections of the particles was measured in the same manner as in the case of the positive electrode active material (A). As a result, no difference in the concentration of each of Mg, Ti and Al was observed between the surface portion and the core portion.

With regard to the contents of the metal elements  $M^2$ , the molar contents of Mg, Ti and Al in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

### Production of Negative Electrode

As negative electrode active materials, 70 parts by weight of a graphite type carbonaceous materiel (a) (purity: 99.9% or more, average particle size: 18  $\mu$ m, d<sub>002</sub>: 0.3356 nm, size of the crystallite in the c axis direction (Lc): 100 nm, R value of the Raman spectrum: 0.18) and 30 parts by weight of a graphite type carbonaceous material (b) (purity: 99.9% or more, average particle size: 21  $\mu$ m, d<sub>002</sub>: 0.3363 nm, size of the crystallite in the c axis direction (Lc): 60 nm, R value of the Raman spectrum: 0.11) were mixed. Then, 98 parts by weight

of the graphite mixture, 1 part by weight of carboxymethyl-cellulose and 1 part by weight of a styrene-butadiene rubber were mixed in the presence of water to prepare a paste containing negative electrode mixture. This paste was uniformly applied to both surfaces of a negative electrode current collector made of a strip-form copper foil having a thickness of 10 µm, and then dried to form negative electrode mixture layers. The resultant sheet was pressed with a roller until the density of the negative electrode mixture layers became 1.75 g/cm³. The resultant sheet was then cut out in a predetermined size. Thereafter, a lead member made of nickel was welded to the cut piece to form a sheet-form negative electrode.

## Preparation of Nonaqueous Electrolytic Solution

An amount of LiPF<sub>6</sub> was dissolved in a mixed solvent of methylethyl carbonate, diethyl carbonate and ethylene carbonate mixed at a volume ratio of 1.5:0.5:1 to attain a concentration of 1.2 mol/L. To this solution, 3% by weight of fluorobenzene, 0.2% by weight of biphenyl, 0.5%; by weight  $^{20}$  of propanesulfone, 10% by weight of  $\rm C_4F_9OCH_3$ , and 3% by weight of vinylene carbonate were added to prepare a non-aqueous electrolytic solution.

#### Production of Nonaqueous Secondary Battery

The positive electrode and the negative electrode were spirally wound with interposing, therebetween, a separator made of a microporous polyethylene film (porosity: 53%, tensile strength in MD:  $2.1 \times 10^8$  N/m<sup>2</sup>, tensile strength in TD: 30 0.28×10<sup>8</sup>N/m<sup>2</sup>, thickness: 16 μm, gas permeability: 80 seconds/100-mL, thermal shrinkage factor after being kept at 105 CC for 8 hours: 3%, puncture strength: 3.5 N (360 g)), to form an electrode body having a spiral structure. Thereafter, the electrode body was pressed to form a flat-shaped electrode 35 body and inserted into a box-shaped battery case made of an aluminum alloy. The positive and negative lead members were welded and a cover plate was laser welded to the edge portion of an opening of the battery case. Then, the nonaqueous electrolytic solution prepared in the above was poured 40 into the battery case through an inlet made in the cover plate. The nonaqueous electrolytic solution was sufficiently infiltrated into the separator and the like. Thereafter, the battery was partially charged, and gas generated during the partial charging was discharged. Then, the inlet was sealed up to 45 make the battery airtight. Thereafter, the battery was charged and aged to yield a rectangular nonaqueous secondary battery having a structure as shown in FIGS. 2A and 2B and an external appearance as shown in FIG. 3, and a width of 34.0 mm, a thickness of 4.0 mm, and a height of 50.0 mm.

Here, the battery shown in FIGS. 2A, 2B and 3 will be explained. The positive electrode 1 and the negative electrode 2 are spirally wound with interposing the separator 3 therebetween, as described above, and the spirally wound electrode body is pressed in a flat form to form the electrode laminate 6 baving a flat spiral structure. The laminate 6 together with a nonaqueous electrolytic solution is contained in the box-shaped battery case 4. For simplicity, in FIG. 2, metal foils as current collectors used to form the positive electrode 1 and the negative electrode 2, and the electrolytic solution are not 60 depicted.

The battery case 4 is made of an aluminum alloy, and constitutes a main part of the exterior package of the battery. This battery case 4 also functions as a positive electrode terminal. The insulator 5 made of a polytetrafluoroethylene 65 sheet is arranged on the inside bottom of the battery case 4. The positive electrode lead member 7 and the negative electrode

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trode lead member 8 connected to one end of the positive electrode 1 and that of the negative electrode 2, respectively, are taken out from the electrode laminate 6 having the flat spiral structure. The terminal 11 made of stainless steel is attached to the cover plate 9 made of aluminum for closing the opening of the battery case 4 with interposing the insulation packing 10 made of polypropylene therebetween. The lead plate 13 made of stainless steel is attached to this terminal 11 with interposing the insulator 12 therebetween.

The cover plate 9 is inserted into the opening of the battery case 4, and their joining portions are welded to each other, thereby closing the opening of the battery case 4 to make the interior of the battery airtight. In the battery shown in FIGS. 2A and 2B, the inlet 14 for pouring the electrolytic solution is made in the cover plate 9, and the inlet 14 is welded and sealed up by, for example, laser welding, with inserting a sealing member (not shown). In this way, the air-tightness of the battery is kept. Accordingly, in the case of the battery shown in FIGS. 2A, 2B and 3, the electrolytic solution pouring inlet 14 is actually composed of the inlet 14 and the sealing member, but the inlet 14 is illustrated as such without a sealing member in order to make the figure simple. The explosion-proof vent 15 is made in the cover plate 9.

In the battery 1 of Example 1, the positive electrode lead member 7 is directly welded to the cover plate 9, whereby the combination of the battery case 4 and the cover plate 9 functions as a positive electrode terminal. The negative electrode lead member 9 is welded to the lead plate 13, and the negative electrode lead member 8 and the terminal 11 are made electrically conductive through the lead plate 13, whereby the terminal 11 functions as a negative electrode terminal. However, the functions of the positive and negative electrodes may be reversed in accordance with the material of the battery case 4, etc.

FIG. 3 is a perspective view schematically illustrating the external appearance of the battery shown in FIGS. 2A and 2B. FIG. 3 shows that the above-mentioned battery is a rectangular battery. Thus, FIG. 3 schematically shows the battery, and depicts the specific elements out of the constituting elements of the battery.

# Example 2

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that fluorobenzene and  $C_4F_9OCH_3$  were not used.

# Example 3

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that propanesul fone was not used.

# Example 4

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive electrode active material (A) was changed to LiCo<sub>0.9988</sub>Mg<sub>0.0008</sub>Ti<sub>0.0004</sub>O<sub>2</sub> (average particle size: 12 μm), and the positive electrode active material (B) was changed to LiCo<sub>0.9964</sub>Mg<sub>0.0024</sub>Ti<sub>0.0012</sub>O<sub>2</sub> (average particle size: 5 μm). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.79 g/cm<sup>3</sup>. With regard to the contents of the metal elements M<sup>2</sup>, the molar contents of Mg and Ti in the positive electrode active material (B) were 3

times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

#### Example 5

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the weight ratio of the positive electrode active material (A) to the positive electrode active material (B) was changed to 90:10. The density of the positive electrode mixture layers (positive electrode) after 10 pressing was 3.75 g/cm<sup>3</sup>.

#### Example 6

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that a mixture of the positive electrode active material (A) consisting of LiCo $_{0.998}$ Mg $_{0.0008}$ Ti $_{0.0004}$ Al $_{0.0008}$ O $_{2}$  (average particle size: 12 µm), and the positive electrode active material (B) consisting of LiCo $_{0.994}$ Mg $_{0.0024}$ Ti $_{0.0012}$ Al $_{0.0024}$ O $_{2}$  (average particle size: 5 µm) in a weight ratio of 50:50 was used as a positive electrode active material. The density of the positive electrode mixture layers (positive electrode) after pressing was 3.76 g/cm³. With regard to the contents of the metal elements M², the molar contents of Mg, Ti and Al in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

#### Example 7

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive elecactive material (A) was changed  $LiCo_{0.998}Mg_{0.0008}Ti_{0.0004}Sn_{0.0008}O_2$  (average particle size:  $^{35}$ 14 µm) and the positive electrode active material (B) was changed to  $\mathrm{LiCo}_{0.994}\mathrm{Mg}_{0.0024}\mathrm{Ti}_{0.0012}\mathrm{Sn}_{0.0024}\mathrm{O}_2$  (average particle size: 6 µm). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.76 g/cm<sup>3</sup>. With regard to the contents of the metal elements M<sup>2</sup>, 40 the molar contents of Mg, Ti and Sn in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

## Example 8

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive elecactive material (A) was changed  $\text{LiCo}_{0.998}\text{Mg}_{0.0008}\text{Zr}_{0.0004}\text{Al}_{0.0008}\text{O}_2$  (average particle size: 13 µm), and the positive electrode active material (B) was changed to  ${\rm LiCo_{0.994}Mg_{0.0024}Zr_{0.0012}Al_{0.0024}O_{2}}$  (average particle size: 6 µm). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.8 55 g/cm<sup>3</sup>. With regard to the contents of the metal elements M<sup>2</sup>, the molar contents of Mg, Zr and Al in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

#### Example 9

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive electrode active material (A) was changed to  $\text{LiCo}_{0.998} \text{Mg}_{0.0008} \text{Ge}_{0.0004} \text{Al}_{0.0008} \text{O}_2 \text{ (average particle size: }$ 

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12  $\mu$ m) and the positive electrode active material (B) was changed to LiCo<sub>0.994</sub>Mg<sub>0.0024</sub>Ge<sub>0.0012</sub>Al<sub>0.0024</sub>O<sub>2</sub> (average particle size: 6  $\mu$ m). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.79 g/cm<sup>3</sup>. With regard to the contents of the metal elements M², the molar contents of Mg, Ge and Al in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

## Example 10

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive electrode active material (A) was changed to LiCo<sub>0.334</sub>Ni<sub>0.33</sub>Mn<sub>0.33</sub>Mg<sub>0.0024</sub>Ti<sub>0.0012</sub>Al<sub>0.0024</sub>O<sub>2</sub> (average particle size: 5 µm). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.72 g/cm<sup>3</sup>. With regard to the contents of the metal elements M<sup>2</sup>, the molar contents of Mg, Ti and Al in the positive electrode active material (B) were 3 times larger, 3 times larger and 3 times larger, respectively, than those in the positive electrode active material (A).

#### Example 11

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that this positive electrode active material (A) was changed to LiCo $_{0.9988}$ Mg $_{0.0008}$ Ti $_{0.0004}$ O $_2$  (average particle size: 12 µm), and the positive electrode active material (B) was changed to LiCo $_{0.9988}$ Mg $_{0.0008}$ Ti $_{0.0004}$ O $_2$  (average particle size: 5 µm). The density of the positive electrode mixture layers (positive electrode) after pressing was 3.79 g/cm³. With regard to the contents of the metal elements M², the molar contents of Mg and Ti were the same in the positive electrode active materials (A) and (B).

# Example 12

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that fluorobenzene was not used.

# Example 13

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that methylethylenesulfate was used in place of propanesulfone.

## Comparative Example 1

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that only  $\text{LiCo}_{0.998}\text{Mg}_{0.0008}\text{Ti}_{0.0004}\text{Al}_{0.0008}\text{O}_2$  (average particle size:  $12~\mu\text{m}$ ) was used as a positive electrode active material. The density of the positive electrode mixture layers (positive electrode) after pressing was  $3.7~\text{g/cm}^3$ . This is a comparative example using only a positive electrode active material having a large particle size, that is, the positive electrode active material (A), among the positive electrode active materials used in Example 1.

# Comparative Example 2

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that only

45

20

27

LiCo $_{0.994}$ Mg $_{0.0024}$ Ti $_{0.0012}$ Al $_{0.0024}$ O $_2$  (average particle size: 5 µm) was used as a positive electrode active material. The density of the positive electrode mixture layers (positive electrode) after pressing was 3.6 g/cm $^3$ . This is a comparative example using only a positive electrode active material having a small particle size, that is, the positive electrode active material (B), among the positive electrode active materials used in Example 1.

#### Comparative Example 3

A nonaqueous secondary battery was fabricated in the same manner as in Example 1 except that the positive electrode active material (A) was changed to  $\text{LiCoO}_2$  (average particle size:  $12~\mu\text{m}$ ), and the positive electrode active material (B) was changed to  $\text{LiCoO}_2$  (average particle size:  $5~\mu\text{m}$ ). The density of the positive electrode mixture layers (positive electrode) after pressing was  $3.8~\text{g/cm}^3$ .

#### Comparative Example 4

A nonaqueous secondary battery was fabricated in the same manner as in Comparative Example 1 except that  $C_4F_9OCH_3$  was not used. This is a comparative example using only a positive electrode active material having a large 25 particle size, that is, the positive electrode active material (A) used in Example 1, and no fluorine-containing organic solvent.

#### Comparative Example 5

A nonaqueous secondary battery was fabricated in the same manner as in Comparative Example 1 except that propanesulfone was not used. This is a comparative example using only a positive electrode active material having a large 35 particle size, that is, the positive electrode active material (A) used in Example 1, and no sulfur-containing organic solvent.

With the nonaqueous secondary batteries fabricated in Examples 1-13 and Comparative Examples 1-5, the following properties were evaluated:

Discharge Capacity after Charge-Discharge Cycles

Each of the batteries fabricated in Examples and Comparative Examples was charged at a constant current of  $0.2\,\mathrm{C}$  up to a voltage of 4.2 V or 4.4 (corresponding to 4.3 V and 4.5 V with reference to the potential of lithium), and then charged at 45 a constant voltage until the total charge time reached 8 hours. Subsequently, the battery was discharged at a constant current of 0.2 C down to a voltage of 3.3 V. Thereby, a discharge capacity of the battery was measured. Then, the above chargedischarge cycle was repeated five times, and the discharge capacity after the fifth cycle was used to evaluate the discharge capacity after the charge-discharge cycles. The results are shown in Table 1. In Table 1, the discharge capacity after the charge-discharge cycles obtained with each battery is shown as a relative value in relation to the discharge capacity of the battery of Comparative Example 1 after the charge- 55 discharge cycles, which is "100".

TABLE 1

Discharge Capacity after Charge-Discharge Cycles			60
Example	Charge	e voltage	
No.	4.2 V	4.4 V	
1 2	103 102	140 137	65

28
TABLE 1-continued

Discharge Capacity after Charge-Discharge Cycles				
Example	Charge voltage			
No.	4.2 V	4.4 V		
3	102	137		
4	103	139		
5	103	134		
6	103	135		
7	103	138		
8	103	136		
9	103	138		
10	103	138		
11	102	122		
12	102	138		
13	102	146		
C. 1	100	100		
C. 2	99	104		

The battery of Example 1 was disassembled after it was charged up to 4.4 V, and the surface of the positive electrode was analyzed by X-ray photoelectron spectroscopy (XPS). The contents of the fluorine atom and sulfur atom were 25 atomic % and 5 atomic %, respectively. In the batteries of Examples 2 and 12, the content of the fluorine atom was 11 atomic % and 20 atomic %, respectively.

102

100

102

73

93

90

The content of the sulfur atoms on the surface of the positive electrode of the battery of Example 3 was less than the detection limit, while it was 4% in the battery of Example 13.

Herein, the XPS analysis of elements was carried out with PHI ESCA 5500MC (manufactured by ULVAC-PHI, Inc.) using the Al—K $\alpha$  ray at 400 W. Then, an obtained curve was subjected to peak separation, and an atomic percentage of each atom is a ratio of the peak area for each atom to the total peak area for all the atoms.

Safety Evaluation

C. 3

C. 4

C. 5

Each of the batteries fabricated in Examples and Comparative Examples was charged at a constant current of 0.2 C up to 4.4 V, and then charged at a constant voltage until the total charge time reached 8 hours. Subsequently, in a temperature-controlled bath, the battery was heated at a heating rate of 5° C./min. up to 150° C. and maintained at 150° C. for 10 minutes. During this 10 minute period, the change of the surface temperature of the battery was monitored. The lower surface temperature during heating means better safety at high temperature. The results are shown in Table 2.

TABLE 2

IABLE 2				
Safety Evaluation				
	Example No.	Charge voltage 4.4 V		
	1	155		
	2	158		
	3	156		
	4	160		
	4 5	155		
	6	162		
	7	159		
	8	156		
	9	156		
	10	154		
	11	167		
	12	157		
	13	156		

Safety Evaluation		
Example	Charge voltage	
No.	4.4 V	
C. 1	>170	
C. 2	168	
C. 3	>170	
C. 4	>170	
C. 5	>170	

As can be seen from the results in Tables 1 and 2, the nonaqueous secondary batteries of Examples 1-13 according to the present invention had larger discharge capacities and better charge-discharge cycle characteristics than those of Comparative Examples 1-5, and further, they suffered from less temperature rising in the full-charged or overcharged state, and had excellent safety.

The invention claimed is:

- 1. A nonaqueous secondary battery comprising:
- a positive electrode having a positive electrode mixture layer, a negative electrode, and a nonaqueous electro-

wherein the positive electrode comprises, as active materials, at least two lithium-containing transition metal oxides having different average particle sizes, and the lithium-containing transition metal oxide having the smallest average particle size is a lithium-containing transition metal oxide represented by the formula (1):

$$\operatorname{Li}_{x}\operatorname{M}^{1}_{\nu}\operatorname{M}^{2}_{z}\operatorname{M}^{3}_{\nu}\operatorname{O}_{2} \tag{1}$$

wherein M<sup>1</sup> represents at least one transition metal element selected from Co, Ni and Mn, M<sup>2</sup> represents Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn,  $\mathrm{M}^3$  represents at least  $^{35}$ one element selected from the group consisting of Na, K, Rb, Be, Ca, Sr, Ba, Sc, Y, La, Hf, V, Ta, Cr, Mo, W, Tc, Re, Fe, Ru, Rh, Cu, Ag, Au, B, Ca, In, Si, P and Bi, and x, y, z and v are numbers satisfying the equations respectively:  $0.97 \le x < 1.02$ ,  $0.8 \le y < 1.02$ ,  $0.002 \le z \le 0.05$ , and  $^{40}$ 0≤v≤0.05, and has an average particle size from 2  $\mu$ m to 10 μm, and the lithium-containing transition metal oxide having the largest average particle size is a lithiumcontaining transition metal oxide represented by the formula (2):

$$\operatorname{Li}_{a} \operatorname{M}^{1}{}_{b} \operatorname{M}^{2}{}_{c} \operatorname{M}^{3}{}_{d} \operatorname{O}_{2} \tag{2}$$

wherein M<sup>1</sup>, M<sup>2</sup> and M<sup>3</sup> are the same as defined in the formula (1), and a, b, c and d are numbers satisfying the equations respectively:  $0.97 \le a < 1.02$ ,  $0.8 \le b < 1.02$ , 50  $0.0002 \le c \le 0.02$ , and  $0 \le d \le 0.02$ , and has an average particle size from 5  $\mu$ m to 25  $\mu$ m,

wherein said electrolyte contains a fluorine-containing organic solvent.

wherein the content of Co in the transition metal M<sup>1</sup> of the 55 according to claim 1 comprising the step of: formulae (1) and (2) is from 30% by mole to 100% by

wherein the content of said lithium-containing transition metal oxide having the smallest average particle size in 30

the lithium-containing transition metal oxides is from 5% by weight to 60% by weight,

wherein the content of said lithium-containing transition metal oxide having the largest average particle size in the lithium-containing transition metal oxides is from 40% by weight to 95% by weight, and

wherein an amount of said fluorine-containing organic solvent is 0.1% by weight to 30% by weight based on the whole weight of the electrolyte.

- 2. The nonaqueous secondary battery according to claim 1, wherein the lithium-containing transition metal oxide having the largest average particle size is a lithium-containing transition metal oxide represented by the formula (2) wherein c and d satisfy the following equation, respectively:  $0.0002 \le c < 0.005$  and  $0 \le d < 0.005$ .
- 3. The nonaqueous secondary battery according to claim 2, wherein z in the formula (1) is larger than c in the formula (2).
- 4. The nonaqueous secondary battery according to claim 1, wherein z in the formula (1) is larger than c in the formula (2).
- 5. The nonaqueous secondary battery according to claim 1, wherein, in the formula (1), the metal elements M<sup>2</sup> are Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn; a content of Mg is from 0.1% by mole to less than 2% by mole based on the amount of the transition metal M<sup>1</sup>; when Ti, Zr, Ge or Nb is contained, a total content thereof is from 0.05% by mole to less than 0.5% by mole based on the amount of the transition metal M<sup>1</sup>; and when Al or Sn is contained, a total content thereof is from 0.1% by mole to less than 1% by mole based on the amount of the transition metal M<sup>1</sup>.
- The nonaqueous secondary battery according to claim 1, wherein, in the formula (2), the metal elements M<sup>2</sup> are Mg and at least one metal element selected from the group consisting of Ti, Zr, Ge, Nb, Al and Sn; a content of Mg is from 0.01% by mole to less than 0.5% by mole based on the amount of the transition metal M<sup>1</sup>; when Ti, Zr, Ge or Nb is contained, a total content thereof is from 0.005% by mole to less than 0.3% by mole based on the amount of the transition metal M<sup>1</sup>; and when Al or Sn is contained, a total content thereof is from 0.01% by mole to less than 0.5% by mole based on the amount of the transition metal M<sup>1</sup>.
- 7. The nonaqueous secondary battery according to claim 1, wherein said positive electrode mixture layer has a density of  $3.5 \text{ to } 4.6 \text{ g/cm}^3$ .
- 8. The nonaqueous secondary battery according to claim 1, wherein said electrolyte further contains a fluorine-containing aromatic compound other than said fluorine-containing organic solvent.
- 9. The nonaqueous secondary battery according to claim 1, wherein the fluorine-containing organic solvent is fluoroethylene carbonate.
  - 10. The nonaqueous secondary battery according to claim 1, wherein M<sup>1</sup> represents Co.
  - 11. A method of using a nonaqueous secondary battery
    - charging the battery so that a positive electrode voltage is in a range of 4.35 to 4.6 V with reference to the potential of lithium when the battery is fully charged.